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# Tectono-magmatic controls of post-subduction gold mineralisation during soft continental collision: Late Caledonian gold in the Southern Uplands-Down-Longford Terrane of Britain and Ireland.

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## Abstract

The Southern Uplands-Down-Longford Terrane (SUDLT) within the British Caledonides hosts several economically significant gold occurrences, including a 45 km-long gold trend in central Ireland that includes the >1 M Oz Au deposit at Clontibret. However, the region is relatively underexplored for gold and the well constrained geology and tectonic history of the terrane provides a firm geological framework for investigating the roles of tectonic, magmatic and metamorphic processes in gold mineralisation during the transition from subduction to soft continental collision. The SUDLT is an Ordovician-Silurian fore-arc accretionary complex that evolved into a foreland basin fold-and-thrust-belt during soft continental collision in earliest Devonian times and is comparable to other Phanerozoic terranes that host orogenic gold deposits globally. Gold mineralisation in the SUDLT exhibits similarities with orogenic, intrusion related gold systems and post-subduction porphyry gold deposits. Gold is predominantly a lattice constituent of arsenopyrite and pyrite occurring within quartz veins and disseminated within structurally controlled phyllic and propylitic potassic alteration haloes within and around intrusions and associated with quartz veins remote from known intrusions. Fluid inclusion data indicate that the gold was deposited from a low salinity mesothermal (~330°C) carbonic fluid of mixed magmatic-metamorphic origin consistent with Caledonian orogenic conditions. Gold mineralisation occurred at shallow crustal depths ~≤5 km, and exhibits a broad spatial association with major NE-SW-trending Caledonoid shear-zones (D<sub>1</sub>). Economically significant gold mineralisation in the SUDLT is most commonly hosted by cross-cutting transverse ~NW-SE- and ~N-S-trending fractures (D<sub>3</sub>) constrained to between 418 and 410 Ma that cut anchizone-epizone facies volcanoclastic turbiditic metasedimentary host rocks. The mineralised D<sub>3</sub> structures in places cut, and in other places

are cut by broadly coeval polyphase diorite and granodiorite intrusions. Gold mineralisation was associated with the early, dioritic I-type metaluminous oxidised magmatic phase of Trans Suture suite magmatism that straddles the Iapetus Suture but predates the later emplacement of S-type granitic magma. The host structures were subsequently reactivated during Late Palaeozoic, Mesozoic and Cenozoic times and host younger base metal deposits (Pb-Zn, Cu, Sn, Sb).

Gold in the SUDLT provides a case study for gold mineralisation in soft continental collision zones globally. Mineralisation occurred in latest Silurian to Early Devonian time (~418 and ~410 Ma) following the arrival of the Avalonian continental margin at the subduction trench at ~420 Ma and was coeval with the onset of regional transtension, post-subduction slab break-off and lithospheric mantle delamination, and K-lamprophyric and mafic to intermediate calc-alkaline magmatism. The transition from convergence to transtension, post-subduction slab delamination and lamprophyric and calc-alkaline magmatism, increasingly recognised as common processes during soft continental collision, provide the critical elements of the mineralising system: a metasomatically fertilised mantle source; transient geodynamics; and favourable lithospheric architecture for the effective rapid transfer of mass and heat energy from subcrustal to upper crustal levels. The inherently thin crust and relatively low degree of exhumation within soft continental collision zones favour the preservation of mineral deposits in the upper crust.

## 1 Introduction

Fore-arc accretionary complexes emplaced within Phanerozoic orogenic belts host a significant proportion of gold deposits globally, predominantly classified as orogenic gold deposits (Böhlke, 1982; Groves et al., 1998). Examples include Ballarat-Bendigo in the Lachlan Fold Belt, Tasman Orogen, eastern Australia; Goldenville and Beaver Dam in the Meguma Terrane, Appalachian Orogen, Nova Scotia; Reefton in the Buller Terrane, South Island, New Zealand; Muruntau in the southern Tien Shan, Central Asia, and Vasilkovskoye in the Kipchak Arc, Kazakhstan (Goldfarb et al., 2001). However, many comparable deposits in this setting are classified as intrusion related gold systems ((IRGS) e.g. Fort Knox, Alaska; Salave, Spain; Mokrsko, Czech Republic and Vasilkovskoe, Kazakhstan; Thompson et al., 1999), and the more recently recognised postsubduction porphyry gold and epithermal types (e.g. Çöpler, Central Turkey; Sari Gunay, northwest Iran; Richards, 2009). The classification of IRGS's and their differentiation from orogenic gold deposits is a long standing problem due to many overlapping characteristics and shared geodynamic settings and conditions of formation (Groves et al., 1998; Hart, 2007; McCuaig and Hronsky, 2014; Thompson et al., 1999; Tomkins, 2013). The problem is compounded by an ongoing debate over metamorphic versus magmatic sources of gold, hydrothermal fluids and sulphur in orogenic deposits (Phillips and Powell, 2009; Pitcairn et al., 2006; Tomkins, 2013). Fluid inclusion and stable isotope data for individual orogenic gold deposits are commonly ambiguous, or indicate mixing between metamorphic and magmatic fluid sources and/or equilibration with country rocks (Oberthuer et al., 1996; Phillips and Powell, 2009; Steed and Morris, 1997; Tomkins, 2013) e.g. Round Hill, New Zealand (de Ronde et al., 2000), Loulo, Mali (Lawrence et al., 2013), Ashanti Belt, Ghana (Mumin et al., 1996; Treloar et al., 2014). Some studies indicate a magmatic origin for the mineralising fluids (e.g. Massawa, Senegal; Treloar et al., 2014) while others suggest fluid of purely metamorphic origin (e.g. Otago and Alpine Schists, New Zealand; Pitcairn et al., 2006). More recently it has been recognised that some gold deposits in post-subduction (i.e. collisional) orogenic settings closely resemble porphyry Au deposits that are generally understood to have formed in shallow sub-volcanic environments in the fore-arc regions of active magmatic arcs (McCuaig and Hronsky, 2014; Richards, 2009; Richards et al., 2006; Sillitoe and Thompson, 1998). The challenge of establishing a general model for gold deposits in orogenic belts has been compounded by the challenges of unravelling the intrinsically

complex geodynamic histories of such regions (Groves et al., 2003). Caledonian gold mineralisation in the Southern Uplands-Down-Longford Terrane (SUDLT) in southern Scotland and Ireland (Figure 1) is relatively poorly explored and has been interpreted in terms of both orogenic (Goldfarb et al., 2001; Lusty et al., 2012) and porphyry gold models (e.g. Boast et al., 1990; Brown et al., 1979; Charley et al., 1989; Duller et al., 1997; Leake et al., 1981; Steed and Morris, 1997). The SUDLT provides an exceptionally well-studied example of a Phanerozoic accretionary complex (together with subjacent penecontemporaneous foreland basin fold-and-thrust belt; Stone, 2014) and a firm geological framework in which to study geodynamic controls and mineralising processes in orogenic settings and sources of heat, fluids and metals.

Detrital gold in drainage sediment is widespread in the SUDLT and more than 25 000 Oz of alluvial gold was extracted historically from the Leadhills-Wanlockhead mining district alone prior to 1876 (Figure 1; Gillanders, 1976). However, no bedrock source of these alluvial deposits has yet been identified. Gold concentrations in bedrock are recorded from eleven other areas of the terrane (Figure 1) and include an economically important 45 Km-long gold trend in Central Ireland that incorporates the Clontibret deposit, which is estimated to contain at least 1 m Oz Au (Cruise and Farrell, 1993). Grades in excess of 50 ppm/m are reported from Fore Burn in southwest Scotland (Charley et al., 1989) and 4.85 ppm Au over 10 m from Moorbrock Hill in southwest Scotland (Beale, 1984). Several authors have investigated the origin of gold mineralisation, mineralising fluids and metals for individual deposits in the terrane using isotopic, geochemical and fluid inclusion data (Duller P.R, 1987; Duller et al., 1997; Lowry, 1991; Lowry et al., 1997; Naden and Caulfield, 1989; Samson and Banks, 1988; Steed and Morris, 1997). An overview of gold mineralisation in the SUDLT is given by (Lusty et al., 2012). The SUDLT is also host to significant lead-zinc and antimony vein deposits related to later hydrothermal events and in places occupying the same lodes as gold.

Here we synthesise the available data for gold mineralisation and review the regional evidence for the nature, conditions, timing and geodynamic context of gold mineralisation within the SUDLT. We find that gold mineralisation occurred at shallow crustal levels and low metamorphic grade during a period of soft collision and postsubduction lithospheric mantle delamination accompanied by regional transtension, high heat flow and polyphased bimodal K-lamprophyric and granitoid magmatism (Leake et al., 1981; Miles et al., 2016). We assess the sources of magma, mineralising fluids, sulphur and gold and find that gold mineralisation

was related to a relatively early phase of hydrous, metaluminous, oxidised, dioritic, I-type magmatism. This magma had a metasomatically hydrated and fertilised subcrustal source and carried gold and sulphur from depth, to be released into an exsolved magmatic-hydrothermal fluid phase at shallow levels, similar to porphyry Au and intrusion-related gold systems (IRGSSs). Magmatic-hydrothermal fluid mixed with fluid derived by low grade metamorphic dewatering of the country rocks. Slab break-off, lithospheric mantle delamination and the transition from convergence to transtension combined with postsubduction magmatism are likely to be inherent to the processes of soft continental collision. Furthermore, through these processes soft continental collision provides the critical elements of the mineralising system, enabling sufficient flux of mass and energy to form and preserve gold deposits at shallow crustal levels in orogenic settings (McCuaig and Hronsky, 2014).

## 2 Geological setting

The SUDLT has an outcrop area of around 20,000 Km<sup>2</sup> in the British Isles (Figure 1). The tectonostratigraphy of the SUDLT is well documented and discussed in detail elsewhere (Floyd, 2001; Leggett et al., 1979a; McKerrow, 1987; Stone, 2014). The succession comprises very low metamorphic grade (anchizone-epizone) meta-volcaniclastic greywacke turbidites deposited in a deep marine basin over a period of ~75 Ma in Ordovician and Silurian times from 495 to 420 Ma (Anderson, 2004; Floyd, 2001; Oliver, 1978; Oliver et al., 2003). The terrane is bounded to the north by the Southern Uplands Fault and to the south by the buried Iapetus Suture (Figure 1), which separates Laurentian from Avalonian basement along the Solway Line (Leggett et al., 1979b; Phillips et al., 1976). Avalonian crust to the south of the Suture is represented by the Lakesman Terrane in northern England, the Isle of Man and central-eastern Ireland. The stratigraphy of the SUDLT is dissected by numerous ~NE-SW-striking faults, roughly subparallel to the strike of bedding (Figure 2). These were originally low-angle thrust faults that developed within a fore-arc accretionary complex and resulted in top-to-the-south thrust imbrication, stratigraphic repetition and thickening (Anderson, 2001; Mitchell and McKerrow, 1975). The major strike-parallel faults demarcate numerous fault-bounded tectonostratigraphic units or 'tracts' (Craig and Walton, 1959). Bedding predominantly youngs to the NW within each tract (Craig and Walton, 1959). However, progressively younger units crop out from northwest to southeast at the terrane scale

(Anderson and Cameron, 1979; Lapworth, 1878; Peach et al., 1899) due to top-to-the-south thrust imbrication (Anderson and Cameron, 1979; Stone et al., 1987). Within each tectonostratigraphic unit hemipelagic black pyritic argillite, the Moffat Shale, passes stratigraphically upwards into a succession of turbiditic greywacke beds (Leggett, 1979; Leggett, 1980; Leggett et al., 1979a). The age of the Moffat Shale and the oldest overlying turbidites is, in general, diachronous across the terrane, becoming younger from northwest to southeast (Leggett et al., 1982; Leggett et al., 1979a; McKerrow et al., 1977), consistent with southeastward progradation of trench-fill sediments derived from the accretionary complex over northwest subducting Iapetus oceanic lithosphere (Mitchell and McKerrow, 1975). The terrane has traditionally been divided into three 'belts' (Anderson, 2001; Anderson, 2004; Stone, 2014). The Northern Belt is dominantly composed of Ordovician turbidites, mainly greywackes of Caradoc to Ashgill age (458-444 My) and is separated from the Central Belt by the Orlock Bridge Fault (Figure 1). The Central Belt is composed mainly of Llandovery greywackes and is separated from the mainly Wenlock age greywackes of the Southern Belt by the Laurieston Fault in Scotland and its continuation in Ireland as the Cloughy Fault (Figure 1; Anderson and Cameron, 1979; Oliver et al., 2003).

Swarms of mid-Silurian to mid-Devonian, mainly calc-alkaline lamprophyric and felsic dykes and several large granitoid 'stitching' plutons ( $408 \pm 2$  to  $395 \pm 2$  Ma; Brown et al., 2008; Halliday et al., 1980; Stephens and Halliday, 1984; Thirlwall, 1988) intrude the turbiditic metasedimentary rocks (Pidgeon and Aftalion, 1978; Read, 1926; Rock et al., 1986). Igneous rocks are described separately below.

To the southwest, in central Ireland, the terrane is buried beneath unconformably overlying shallow-marine shelf carbonates of Lower Carboniferous age (Figure 1; George, 1958; Guion et al., 2000; Lewis and Couples, 1999; Mitchell, 2004). A much thinner Carboniferous succession comprising volcano-sedimentary rocks is preserved within narrow NW-SE and N-S trending fault-bounded half-graben within the terrane (Figure 1). Within these basins Carboniferous rocks, if present, are succeeded by thicker Permo-Triassic volcano-sedimentary successions composed of basic lavas and terrestrial red sandstone (Anderson et al., 1995; Caldwell and Young, 2013; Coward, 1995; Pringle and Richley, 1931).

Long-standing debate over the origin of the terrane has recently been largely resolved (Stone, 2014). Ordovician rocks, predominantly cropping-out in the Northern Belt of the SUDLT, represent a fore-arc subduction-accretion complex as originally proposed by Dewey

(1969) and Mitchell and McKerrow (1975). The sediments apparently did not sample any coeval magmatic arc, indicating probable amagmatic subduction (Miles et al., 2016; Phillips et al., 2003). Silurian rocks of the Central and Southern Belts of the SUDLT were deposited following the arrival of the Avalonian continental margin at the subduction trench at ~430 Ma and represent a foreland fold and thrust belt (Hutton and Murphy, 1987; Stone, 2014; Stone et al., 1987). Clockwise transection of folds by cleavage indicates a change from orthogonal accretion to transpressional deformation at this time (Anderson, 1987; Dewey and Strachan, 2003). Very low metamorphic grade and predominantly moderate brittle deformation styles indicate a 'soft' continental collision (Stone, 2014). Deposition in the SUDLT was terminated by the end of Wenlock times (422 Ma) and followed by magmatism, uplift and emergence (Anderson, 1987; Dewey and Strachan, 2003; Kemp, 1987; Miles et al., 2016; Stone, 2014). Early Devonian exhumation and erosion of up to 20 km was accompanied by terrestrial deposition of the Old Red Sandstone Group within transtensional basins controlled by sinistral strike-slip and normal faults (Anderson et al., 1995; Bluck, 1984; Coward, 1995; Dewey and Strachan, 2003; Leeder, 1982). Transtension between 420 and 405 Ma was accompanied by the onset of lamprophyric and calc-alkaline magmatism (Miles et al., 2016). It is important to emphasise that Caledonian calc-alkaline magmatic rocks of northern Britain and Ireland post-date final closure of Iapetus by up to 40 Ma (Miles et al., 2016).

### 3 Structure

The structure of the terrane is remarkably uniform (Figure 2): The 'Caledonoid' structural grain ( $D_1$ ) is defined by the predominantly NE-SW strike of the generally subvertical to steeply dipping turbidite beds, exhibiting tight to isoclinal asymmetrical folds ( $F_1$ ), strike-parallel faults and subparallel slaty cleavage ( $S_1$ ) best developed in fine-grained argillaceous lithologies (Figure 2; Anderson, 2001; Barnes et al., 1987; Stringer and Treagus, 1980).  $D_1$  faults are subvertical to steeply-dipping back-rotated thrusts that predominantly downthrow to the south and have been reactivated by strike-slip.  $F_1$  folds are generally tight to isoclinal, predominantly highly asymmetrical upright folds with ~NE-SW trending axes having variable plunge and exhibit ~SE-vergence (Figure 2; Anderson, 2004; Barnes et al., 1987; Barnes et al., 2008; Stone et al., 2012; Stringer and Treagus, 1980).



D<sub>1</sub> structures reflect deformation due to tectonic development of the accretionary complex during northwards underthrusting and accretion of Iapetus oceanic lithosphere in an active forearc setting (Dewey and Strachan, 2003). Argillaceous rocks of the Moffat Shale Group acted as the principal decollement during D<sub>1</sub> thrusting (Barnes et al., 1995; Leggett et al., 1979a). In the younger southeasterly units S<sub>1</sub> cleavage obliquely transects F<sub>1</sub> folds with a clockwise sense by  $\sim 20^\circ$  reflecting a progressively non-orthogonal relationship between the principal compressive stress and the orientation of bedding, indicating a change from orthogonal to oblique subduction during arrival of the Avalonian margin at the subduction trench in Early Silurian time (Anderson, 2004; Stone et al., 2012; Stringer and Treagus, 1980; Anderson and Cameron, 1979).

The Moniaive Shear Zone (MSZ) in Scotland and its continuation, the Slieve Glah Shear Zone (SGSZ) in Central Ireland (Figure 1), is a zone up to 5 km wide of enhanced ductile deformation subparallel to the regional Caledenoid structural grain. It dips steeply NW and marks the boundary between the Northern and Central Belts (Anderson and Oliver, 1986; Oliver, 1978; Phillips et al., 1995). The MSZ/SGSZ is cut by and locally bounded on its northern side by the Orlock Bridge Fault (OBF). In central Ireland the SGSZ hosts a 45 km long gold trend that includes the Clontibret deposit (Figure 1; Cruise and Farrell, 1993; Lusty et al., 2012). Within the shear zone, Silurian greywackes of the Gala Group exhibit pervasive linear and planar fabrics including mylonite, extensional crenulation cleavage and stretching lineation with a consistently sinistral sense of shear (Phillips et al., 1995; Stone, 1996). Sinistral deformation on the MSZ post-dates D<sub>1</sub> and probably represents strike-slip reactivation of an over-steepened major tract-bounding fault during Wenlock times (Barnes et al., 1995). The superposition of brittle structures upon the ductile fabrics of the shear zone indicates that sinistral strike-slip deformation accompanied progressive exhumation through the brittle-ductile transition zone (Phillips et al., 1995).

F<sub>1</sub> folds and S<sub>1</sub> cleavage are deformed locally by gently to moderately inclined open to close folds (D<sub>2</sub>) with gently to moderately inclined axial surfaces and associated crenulation cleavage (Barnes et al., 1987). D<sub>2</sub> structures are generally weakly developed (Figure 2).

Two conjugate pairs of steeply inclined faults and spaced fracture cleavage representing D<sub>3</sub> are developed transverse to the regional NE-SW trending D<sub>1</sub> structural grain (Figure 2). These structures strike NW-SE (110-150°) and  $\sim$ N-S (170°-030°; Figure 3; Stone et al., 1995; Stone et al., 2012) and host most of the metalliferous lodes e.g. Pb-Zn veins at

Leadhills-Wanlockhead and Whitespots; Sb-Au lodes at Clontibret, Hare Hill and Glendinning (Figure 1, 2, 3, 4). Some of the transverse faults exhibit breccia zones ~1m thick with stockworks and vein-filled fractures (Anderson, 1987; Moles and Nawaz, 1996; Morris, 1984; Temple, 1956). Dextral subhorizontal slickensides are dominant on the NW-SE striking faults whereas sinistral subhorizontal slickensides are dominant on the N-S faults (Figure 3). Limited lateral offset is exhibited across transverse D<sub>3</sub> faults. Steeply plunging D<sub>3</sub> folds of D<sub>1</sub> cleavage are developed adjacent to D<sub>1</sub> and D<sub>3</sub> faults, indicating that D<sub>3</sub> sinistral strike-slip also reactivated strike-parallel D<sub>1</sub> thrusts (Anderson and Cameron, 1979; Stone et al., 2012; Stringer and Treagus, 1980).

#### 4 Timing of deformation

Minor intrusions cutting the Slieve Glah Shear zone in Ireland indicate that compressional D<sub>1</sub> deformation on the Shear Zone occurred prior to c. 400 Ma (Anderson and Oliver, 1986). In Country Down and Galloway a change from foliated to unfoliated lamprophyre dykes together with clockwise-transecting cleavages indicate that orthogonal accretion switched to sinistral transpression at around 400 Ma (Anderson, 1987; Barnes et al., 2008; Dewey and Strachan, 2003; Miles et al., 2016; Rock et al., 1986; Stone, 1995). A maximum age for the initiation of the transverse strike-slip faults (D<sub>3</sub>) is provided by broadly contemporaneous lamprophyre dykes in Galloway dated between 400 and 418 Ma (K-Ar method) that are in some cases cut by, and in others contained by the faults (Anderson, 1987; Rock et al., 1986). In places the dykes are brecciated and exhibit the same sense and magnitude of displacement as the country rocks, indicating pre-kinematic intrusion (Anderson, 1987; Rock et al., 1986). In other cases, the dykes follow D<sub>3</sub> faults, cut breccia zones and exhibit apparent displacements in the opposite sense to the country rocks indicating post-kinematic intrusion (Anderson, 1987). The dykes are best exposed on the Galloway coast near Kirkudbright where they intrude 420 My old Llandovery-Wenlock country rocks and are not found intruding the Criffel Pluton dated at 410 ± 6 Ma (Miles et al., 2014).

Within the contact metamorphic aureole of the Fleet pluton cordierite porphyroblasts have overgrown the early mylonitic fabric D<sub>1</sub> of the Moniaive Shear Zone (Phillips et al., 1995). However, the porphyroblasts have been deformed by subsequent reactivation of the shear

zone. Recent U/Pb zircon geochronological evidence for the Fleet pluton indicates that the reactivation of the Moniaive Shear Zone occurred between the two intrusive phases at 410 and 387 Ma (Miles et al., 2014). In northern England and Wales clockwise-transecting transpressive regional cleavages mark the onset of Acadian transpression at 404 Ma (Miles et al., 2016). The locations and geometries of the granitoid plutons appear to be influenced by D<sub>1</sub> and D<sub>3</sub> structures. For example, the Cairnsmore of Carsphairn (pluton 410.4 ± 4 Ma, Rb-Sr method; Thirlwall, 1988) crops out near the intersection of the Leadhills fault and the NNW-SSE trending Luke's Stone Fault. Strong ~N-S elongation (Figure 1) of the Loch Doon Plutonic Complex (408 ± 2 Ma, K-Ar method; Stephens and Halliday, 1984) together with locally developed internal ~N-S foliation and asymmetrical vertical drag folds in contact metasedimentary country rocks indicates syn-magmatic N-S sinistral shear (Leake et al., 1981).

Due to extensive reactivation of the transverse D<sub>3</sub> strike-slip the minimum age of fault motion is not known. However, regional evidence indicates that reactivation of D<sub>3</sub> faults occurred in Carboniferous, Permian and Palaeogene times under extensional regional stress fields leading to further deposition within existing fault-bounded volcano-sedimentary basins e.g. the East Irish Sea, Solway Firth, North Channel and Firth of Clyde, Kingscourt, Strangford, Luce/Stranraer, Dumfries, Thornhill, Sanquhar and Langholm (Figure 1; Anderson et al., 1995; Barnes et al., 2008; Caldwell and Young, 2013; Coward, 1995; Floyd et al., 2007; Mitchell, 1992; Oliver et al., 2003; Ruffell and Shelton, 2000; Stone, 1995; Stone et al., 1995). Thicknesses of Permo-Carboniferous volcanosedimentary successions in these basins and contrasts in metamorphic grade across D<sub>3</sub> faults indicate vertical displacements in excess of 2 km (Anderson et al., 1995; Stone et al., 1995). Reactivation of D<sub>3</sub> faults during these extensional events could have remobilised gold. This is supported by evidence from the distribution and character of alluvial gold in the Thornhill area (Leake and Cameron, 1996; Leake et al., 1998) and by atypical oxide-related lode-gold mineralisation in Western Ireland (Lusty et al., 2011). It is likely that regional hydrothermal activity and Pb-Zn-Cu-Ag vein mineralisation was related to this tectonic reactivation (Baron and Parnell, 2005; Wilkinson et al., 1999). Detailed geochronological studies could help clarify the timing of these events and their possible role in gold remobilisation.

## 5 Magmatism

Caledonian igneous rocks in the SUDLT are represented by large calc-alkaline plutonic complexes plus swarms of mafic K-lamprophyres and appinites, monzonite, granodiorite, felsic quartz-porphyry and microgranite dykes (Anderson and Cameron, 1979; Barnes et al., 2008; Brown et al., 2008; Leake et al., 1981; Leake and Cooper, 1983; Miles et al., 2016; Rock et al., 1986). The granitoid plutons of the Southern Uplands belong to the Trans Suture Suite (TSS; Brown et al., 2008; Miles et al., 2014; Miles et al., 2016), that includes all late Caledonian granitoids south of the Highland Boundary Fault (Miles et al., 2016). Rocks belonging to the TSS exhibit similar petrological and geochemical character (Brown et al., 2008; Miles et al., 2016). The TSS spans the buried trace of the Iapetus Suture between Laurentia and Avalonia (Brown et al., 2008). The proportion of Caledonian S-type relative to I-type granitoids generally increases southwards from the Grampian Highlands to the Lakesman Terrane (Brown et al., 2008). The affinity of the Newry Igneous Complex in County Down is less well constrained (Anderson et al., 2016).

Geophysical evidence for buried large plutons in the Tweeddale area suggest that the volume of TSS late Caledonian granitoid intrusions in the SUDLT is likely to be greater than currently estimated (Miles et al., 2016). Magmatism is therefore likely to have had a substantial influence on the thermal regime and the activity of fluids, sulphur and metals in the SUDLT during early Devonian times. Recent U-Pb zircon ages show that the TSS was emplaced between 426 and 387 Ma, broadly coeval with granitoids in the Grampian Highlands and indicating a common origin (Miles et al., 2016). The granitic magmatism was also coeval with emplacement of K-lamprophyre dykes, dated between 400 and 418 Ma (K-Ar method; Anderson, 1987; Rock et al., 1986), and some granitic bodies exhibit lamprophyric enclaves (Brown et al., 2008).

The granitic plutons are generally zoned with older, more mafic dioritic rims and younger, more silicic, granitic cores (Brown et al., 2008; Halliday et al., 1980; Stephens and Halliday, 1984). The outer zones have more metaluminous I-type compositions, whereas the cores have more peraluminous S-type compositions (Brown et al., 2008). For example, Criffel-Dalbeattie is concentrically zoned with outer, early I-type hornblende granodiorite enveloping a core of S-type two-mica granite (Stephens, 1992; Stephens et al., 1985). Cairnsmore of Fleet is a two-mica granite pluton with S-type chemical and isotope characteristics (Brown et al.,

2008). Loch Doon is a zoned I-type dioritic to granitic complex with ferric/ferrous ratio between ~0.66 and 2.88. In each of these plutons isotopic ratios and REE abundances vary systematically from the outer to the inner zones, indicating an increased input of crustal material through time (Brown et al., 2008). Initial  $^{86}\text{Sr}/^{87}\text{Sr}$  ratios (0.705 to 0.708) together with  $\delta^{18}\text{O}$  (8 to 12 ‰) and  $\epsilon\text{Nd}$  values (-3.4 to -0.6) for Southern Uplands plutons indicate the same magmatic source as for the north of England (Brown et al., 2008). Pb isotopes match those of the Ordovician Skiddaw Group in the Lake District, indicating a possible contribution from Avalonian crust (Brown et al., 2008).

The age spectra of the granitic plutons show that magmatism occurred both sides of the suture zone during regional transtension that immediately followed the termination of Iapetus subduction (Miles et al., 2016). Recognition of coeval mantle-derived lamprophyric and crustal S-type granitic melts supports the role of magmatic heat advection in generating anatectic granites (Brown et al., 2008). Isotopic characteristics of the TSS plutons indicate a source in Avalonian crust that was underthrust beneath the Iapetus Suture, including a component derived from pelites of the Skiddaw Group (Miles et al., 2016). The lamprophyres and appinites were probably sourced in sub-continental mantle previously metasomatised by subduction prior to collision (Miles et al., 2016). The most probable tectonic model to explain the postsubduction emplacement of these magmas both sides of the ISZ is southwards propagating delamination of the Avalonian sub-continental lithospheric mantle (Miles et al., 2016). Comparable postsubduction lithospheric delamination has been proposed in the Eastern Mediterranean region on the basis of seismic tomography (van Hinsbergen et al., 2010). The hydrous granitoid magmas most probably indicate a metasomatically hydrated mantle source, likely to be also enriched in sulphide and metals. The metalliferous magmatic sulphide deposit at Talnotry demonstrates that at least some of the more primitive (appinitic) magmas have transported gold, PGE's, Cu and Ni from the lower crust or mantle and have become sulphide-saturated at higher crustal levels. The compositions of the granitic plutons in the SUDLT are comparable to those in the Lachlan Fold Belt, Australia (Chappell and White, 2011) and indicate prospectivity for a range of metalliferous deposit types including Cu (Au, Mo) porphyry and Sn-W skarns (Barton, 1996; Robb, 2009).

Whole rock geochemical data for Caledonian minor igneous intrusions in the Southern Uplands were provided by the British Geological survey and screened to identify ultrapotassic rocks following the parameters used by Muller and Groves (Müller and Groves, 2016). Out of

357 samples 116 fell within the range for ultrapotassic rocks. These results were then plotted on a hierarchical series of tectonic discrimination diagrams following Muller and Groves (2016; Figure 4). The discrimination diagrams use incompatible immobile element ratios in order to minimise interference by the effects of fractionation, alteration, weathering and inter-laboratory differences. The diagrams in Figure 5a, d and e show that none of the samples are within-plate potassic igneous rocks. Figure 5b shows that the samples are of post-collisional and/or continental margin volcanic arc type and not juvenile or mature oceanic arc. Only 6 of the ultrapotassic samples were analysed for cerium and could therefore be plotted on the  $\text{Ce}/\text{P}_2\text{O}_5$  versus  $\text{Zr}/\text{TiO}_2$  diagram Figure 5c to separate post-collisional from continental margin volcanic arc ultrapotassic rocks. Four of the samples fall clearly within the field for continental arc and 2 fall just inside the field for post-collisional arcs. It is important to note that on Figure 5c there is overlap between continental and post-collisional arcs. The ternary diagram in Figure 5f provides a better separation between continental margin and post-collisional arcs and on this diagram, with the exception of one sample, the lamprophyres fall mainly in the post-collisional arc field. This analysis provides geochemical evidence that lamprophyric and coeval TSS granitoid magmatism occurred within a post-collisional arc-setting. This was also the context of gold mineralisation, as indicated by the structural relationships, mineral assemblages and fluid inclusions described below.

## 6 Description of gold mineralisation

Similar sulphide mineral assemblages are exhibited at localities where gold mineralisation is hosted by, or associated with, igneous intrusions and veins that are remote from any known intrusions. At all of the known gold-in-bedrock localities in the SUDLT (Figure 1) auriferous veins are generally <10 cm thick and contain quartz  $\pm$  subordinate carbonate together with sulphides (Allen et al., 1982; Boast et al., 1990; Brown et al., 1979; Duller P.R, 1987; Leake et al., 1981; Steed and Morris, 1986). The sulphides associated with gold are principally arsenopyrite, pyrite and chalcopyrite and occur as veins and disseminations within the veins and sericitised wallrocks. At Clontibret gold grades are highest in the wall rocks (Morris et al., 1986). Native Au grains are generally rare in bedrock throughout the terrane. However, gold has been observed as small inclusions <20  $\mu\text{m}$ , locks <50  $\mu\text{m}$  and fracture fills within brecciated arsenopyrite and pyrite at Fore Burn and Glendinning (Boast et al., 1990;

Charley et al., 1989; Duller et al., 1997) and grains  $<10\ \mu\text{m}$  were recovered from sericitised granodiorite at Hare Hill (Boast et al., 1990). Rare grains of particulate gold generally  $<10\ \mu\text{m}$  were identified within quartz and pyrite but not arsenopyrite at Clontibret (Morris et al., 1986). At none of these localities was the abundance of gold grains sufficient to account for the corresponding gold grades, suggesting that the gold is predominantly sub-microscopic and most probably a lattice constituent of arsenopyrite and pyrite (Boast et al., 1990), i.e. 'refractory ore'.

Gold and geochemical pathfinder element anomalies in soil and bedrock predominantly exhibit  $\sim\text{NE-SW}$  ( $D_1$ ) and  $\sim\text{N-S}$  trending ( $D_3$ ) elongation and occur within metasedimentary rocks and late Caledonian hypabyssal intrusions (Figure 5; Boast et al., 1990; Leake et al., 1981). The anomalies may correspond to zones of simple quartz veining, e.g. at Glenhead and Hare Hill (Leake et al., 1981), or to more complex zones of fracturing, brecciation and fault gouge containing veins and disseminations of quartz-sulphide mineralisation, e.g. at Clontibret, Glendinning, Duns and Moorbrock Hill (Beale, 1984; Duller P.R, 1987; Duller et al., 1997; Morris, 1984; Steed and Morris, 1986).

Whether within, proximal to or remote from igneous intrusions, the gold-mineralised structures are consistently enveloped by distinctive zones of metasomatic phyllic (sericite) alteration with veinlets and patches of disseminated sulphides including auriferous arsenopyrite and pyrite (Figure 7; e.g. Boast et al., 1990; Brown et al., 1979; Duller et al., 1997; Leake et al., 1981; Morris et al., 1986; Steed and Morris, 1986). Fragments of altered and mineralised brecciated wallrocks within fault breccia, e.g. at Clontibret, demonstrate reworking and a polyphase history of fluid flow and deformation of the lode zone (Morris, 1984; Morris et al., 1986). Detailed analysis of the lode zones at Clontibret has revealed a complex paragenetic sequence with six generations of hydrothermal mineralisation in which gold is associated only with phases 4 and 5 (Table 1; Morris et al., 1986). There are no data for the occurrence or paragenetic sequence of gold in veins at Laeadhills-Wanlockhead. However, it has been reported that stream sediment  $\text{Au}$  anomalies are spatially associated with the traces of the  $\text{Pb-Zn}$  veins (Gillanders, 1976; Porteous, 1876) and detailed paragenetic study indicates a likely association between gold and relatively early vein generations within the polyphase sequence (Temple, 1956). Significant anomalous gold concentrations have been identified in felsic dykes in the Leadhills area (Boast and Harris, 1984). Early reports indicate that gold-bearing veins in the LeadhillsWanlockhead area were found within deeply

weathered saprolitic regolith (Porteous, 1876). Complete paragenetic sequences have not been established for the other gold-bearing localities.

The base of an appinite intrusion at Talnotry (Figure 1) exhibits magmatic polymetallic precious metal mineralisation (Power et al., 2004; Stanley et al., 1987) with inclusions of electrum (80% Au) within magmatic chalcopyrite (Power et al., 2004). Compositions and textures exhibited by the pyrrhotite-chalcopyrite assemblage indicate that monosulphide solid-solution crystallization led to enrichment of Ni, Cu, Pt, Pd, Au and As in the residual sulphide liquid (Power et al., 2004). A Cu and Au-rich phase subsequently crystallised to form discordant cross-cutting electrum-bearing chalcopyrite veins (Power et al., 2004). The mineralisation represents a magmatic sulphide cumulate deposit and strongly supports the potential role of magmatic processes in concentrating gold in the SUDLT. Comparable deposits of similar age are found in the Grampian Terrane at Sron Garbh and in the Northern Highlands (Graham et al., 2017). Other localities where gold has been identified in bedrock in the SUDLT, for example Black Stockarton Moor, have been interpreted in terms of magmatic-hydrothermal processes. Mineralisation at Black Stockarton Moor occurs within hydro-fractured and metasomatised rocks immediately above granodiorite sheets within a subvolcanic complex and represents a porphyry Cu (Au) type deposit (Brown et al., 1979). Very similar geochemical relations and hydrothermal alteration assemblages are observed at Glendinning, Fore Burn and Hare Hill and Glenhead indicating a common source of magmatic-hydrothermal gold-mineralising fluids. However, these deposits have not been classed as porphyry-type due to a lack of clear zonation of alteration, structural control of mineralisation or the lack of evidence for a proximal source intrusion (Boast et al., 1990; Charley et al., 1989; Leake et al., 1981; Steed and Morris, 1986).

## 7 Lithological relations

### 7.1 Greywacke

Gold mineralisation in the SUDLT is concentrated in the Northern Belt, i.e. north of the OBF (Figure 1; Lusty et al., 2012). Two exceptions are Black Stockarton Moor and Glendinning, both located in the Southern Belt. However, to date neither of these localities have yielded Au concentrations greater than 0.84 ppm (Brown et al., 1979; Leake et al., 1981). Some units



north of the Northern Belt, e.g. the Portpatrick Formation, contain more andesitic and mafic detritus than those south of the OBF (Floyd, 2001; Stone et al., 1995). In addition, the Northern Belt exhibits a regional relative enrichment in As, Pb and Zn (Stone et al., 1995). The spatial correlation between gold mineralisation and more metalliferous metasedimentary units may indicate that metals were derived from the more mafic-rich greywackes of the Northern Belt or that the greywackes of the Northern Belt were more chemically reactive with metalliferous mineralising fluids than the rocks in the Central and Southern Belts (Lusty et al., 2012). However, gold does not exhibit a strong association with any particular stratigraphic unit within the Northern Belt (Table 2) and significant Au anomalies are found within a range of lithologies including greywacke sandstone, carbonaceous black shale major and minor dioritic, granodioritic and porphyritic intrusions (Boast et al., 1990; Leake et al., 1981; Lowry et al., 1997; Morris et al., 1986; Naden and Caulfield, 1989). Greywacke sandstones hosting gold mineralisation occur at a range of stratigraphic levels from Middle Ordovician to Wenlock. For example, the Llandeilian Red Island Formation hosts the prospect at Glenish and the Hawick Group hosts auriferous As-Sb mineralisation at Glendinning (anonymous, 2015; Morris, 1983; Morris, 1984; Shaw et al., 1995).

## 7.2 Black Shale

Black pyritic carbonaceous shale of the Moffat Shale Group is commonly the locus of D<sub>1</sub> shear zones that exert a primary control on the localisation of gold mineralisation in the terrane (Lusty et al., 2012). The Moffat shale is spatially associated with gold mineralisation in the Leadhills-Wanlockhead area, at Clay Lake and Slieve Glah in Central Ireland and at Moorbrock Hill (Figure 1; anonymous, 2014a; anonymous, 2014c; Boast and Harris, 1984). Discordant ~N-S mineralised faults and fractures within the Moffat shale are intruded by quartz microdiorites immediately south of Glenhead where they host auriferous sulphide bearing quartz veins (Leake et al., 1981). The Moffat Shale forms the main decollement in the Leadhills Imbricate Zone and is likely to be an important host rock, fluid conduit, possible source of sulphur and metals and a structural pathway for gold. In addition, carbon derived from the black shale during deformation and fluid flow is likely to have provided a ligand for gold transport and also created a reducing environment promoting the stability of dissolved gold-sulphide complexes. Sheared black carbonaceous shales commonly host auriferous lodes within Phanerozoic and some Proterozoic orogenic gold deposits globally, e.g. the

489 Ashanti Belt in the Birimian Shield of West Africa (Goldfarb et al., 2001; Groves et al., 2003;  
 490 Oberthuer et al., 1996).

### 491 7.3 Igneous rocks

492 A spatial correlation between metalliferous mineralisation in the SUDLT and the  
 493 outcrops of large plutons has been suggested (Lowry et al., 1997; Naden and Caulfield, 1989)  
 494 but is not ubiquitous and notable exceptions include alluvial gold at Leadhills, Glendinning  
 495 and the Central Irish gold trend including Clontibret (Lusty et al., 2012). However, greywacke  
 496 turbidites are hornfelsed in the vicinity of Clontibret indicating a probable buried intrusion  
 497 and geophysical data support the possibility of unexposed large intrusions here and at  
 498 Glendinning, Stobshiel and Leadhills (Duller et al., 1997; Leake et al., 1996; Morris et al., 1986;  
 499 Steed and Morris, 1986). An association with minor intrusions has also been suggested  
 500 (Brown et al., 1979; Charley et al., 1989; Duller et al., 1997; Leake et al., 1981). The PGE-  
 501 enriched magmatic sulphide cumulate deposit at Talnotry is a clear example of a magmatic  
 502 igneous source of gold and indicates that late Caledonian magmas were capable of  
 503 transporting and concentrating precious metals (Power et al., 2004). Gold anomalies are  
 504 found in the margins of the Loch Doon Pluton (Glenhead Burn; Leake et al., 1981), the  
 505 Cairnsmore of Carsphairn Pluton (Moorbrock Hill; Beale, 1984), the Criffel Pluton (Black  
 506 Stockarton Moor; Brown et al., 1979), the Fleet Pluton (Talnotry; Power et al., 2004), the  
 507 Newry Igneous Complex in County Down (Toal and Reid, 1986), the Priestlaw Pluton in the  
 508 Duns area and at Stobshiel (Figure 1; Naden and Caulfield, 1989; Shaw et al., 1995). In  
 509 addition, relationships between minor intrusions, metasomatic alteration and gold  
 510 mineralisation have been shown at several localities, e.g. Glenhead (Leake et al., 1981), Black  
 511 Stockarton Moor (Brown et al., 1979), Fore Burn (Allen et al., 1982; Charley et al., 1989) and  
 512 Leadhills (Boast and Harris, 1984). At some of the gold-bearing localities intensely K-altered  
 513 felsic intrusions are found, as would be expected for porphyry-gold and intrusion-related gold  
 514 systems (Allen et al., 1982; Berger et al., 2008; Boast et al., 1990; Brown et al., 1979; Charley  
 515 et al., 1989; Leake et al., 1981; Robb, 2005; Rose, 1970; Sillitoe, 1991; Sillitoe and Thompson,  
 516 1998). Antimony-gold mineralisation at Hare Hill, with gold grades up to 5 ppm, is hosted  
 517 within a moderate-scale late Caledonian slightly porphyritic biotite-hornblende granodiorite  
 518 intrusion, ~1.6km in diameter (Figure 4; Boast et al., 1990). The mineralisation occurs as  
 519 discrete structurally controlled lodes corresponding to zones of faulting and veining with

haloes of sericitic alteration and disseminated sulphides (Boast et al., 1990). Gold grades up to 4.85 ppm Au over 10 m are spatially associated with the dioritic margin of the Carsphairn igneous complex that intrudes the Moffat shale within the Leadhills Fault Zone at Moorbrock Hill (Beale, 1984; Dawson et al., 1977). Lamprophyric and felsic porphyritic dykes also crop out locally (Survey, 2016). At Fore Burn (Figure 1), two gold anomalies are identified at surface and in drill core within metasomatised Lower Devonian intermediate to acidic volcanic and subvolcanic intrusive igneous rocks (Allen et al., 1982; Charley et al., 1989). The western anomaly corresponds to a NW-SE-trending lode zone with gold grades up to 50 ppm Au for 90 cm true width (Charley et al., 1989). The mineralised faults cut rhyodacites, tourmaline breccias and intermediate porphyries that exhibit intensive sericite alteration of possible subvolcanic origin and the mineralisation at Fore Burn is interpreted as epithermal (Charley et al., 1989). Fore Burn is located immediately north of the SUF and therefore lies outside of the SUDLT *sensu stricto*. However, Fore Burn exhibits marked similarities to the other auriferous localities in terms of timing, host rocks, structure and mineral assemblages.

Dyke emplacement and associated metasomatic alteration preceded emplacement of the granitoid plutons at Black Stockarton Moor and Glenhead at  $397 \pm 2$  and  $408 \pm 2$  respectively (Rb-Sr isotope method; Brown et al., 1979; Halliday et al., 1980; Leake et al., 1981; Stone and Leake, 1984). At Glenhead, the intensity of hydrothermal alteration in hornfelsed greywacke country rocks does not correlate with distance from the margin of the pluton (Leake et al., 1981). Furthermore, xenoliths of altered country rock are found within the margins of the Loch Doon Plutonic Complex, indicating that metasomatic K-alteration preceded its emplacement (Figure 7; Leake et al., 1981). Two phases of gold mineralisation are identified at Glenhead: 1) Weak disseminated As-Au mineralisation, <0.14 ppm Au is associated with the margins of concordant late Caledonian monzonite dykes (Leake et al., 1981); 2) Higher gold grades, <8.8 ppm Au, are associated with discordant ~N-S veins that cut the metawacke country rocks, minor intrusions and the early-formed dioritic margin of the pluton, indicating that gold mineralisation overlapped in time with the early stages of its emplacement (Figure 7). These ~N-S veins have strong cm-scale post-metamorphic sericitic alteration haloes with disseminated sulphides (Leake et al., 1981). Concordant granodiorite intrusions at Glenhead exhibit no Au-As anomalies. Taken together, these observations indicate that Au mineralisation post-dates the early stages of Caledonian magmatism and metamorphism but pre-dates the later stages of granitoid emplacement (Leake et al., 1981).

However, in detail, relationships between deformation, magmatism and mineralisation are likely to be complex: gold mineralisation is likely to have been polyphased, but broadly coeval with magmatism and deformation.

Greywacke turbidites are intruded by a subvolcanic complex of probable earliest Devonian age at Black Stockarton Moor (Figure 1; Brown et al., 1979). Turbidites immediately above granodiorite sills are hydro-fractured and quartz-veined and exhibit zoned metasomatic phyllic-propylitic alteration comparable to that seen at the other gold bearing localities (Figure 6; Brown et al., 1979). The fractured and metasomatised zones at Black Stockarton Moor have very low grade anomalous As-Au values (Brown et al., 1979). Although gold abundances so far recorded are very low, structural relationships and mineral assemblages are similar to the other localities and indicate comparable timing, P-T-X conditions and tectono-magmatic causes of metasomatism and mineralisation. Mineralisation and alteration at Black Stockarton Moor are most probably the result of the rapid release of magmatic-hydrothermal fluids at a shallow crustal level (Brown et al., 1979; Clarkson et al., 1975; Craig and Walton, 1959; Floyd et al., 2007). The mineralisation is interpreted as porphyry Cu (Au) type (Brown et al., 1979).

Numerous minor felsic porphyritic intrusions are mapped in the Leadhills-Wanlockhead area, spatially associated with alluvial gold concentrations of economic significance (Survey, 2000; Survey, 2016). However, data for gold and pathfinder elements geochemistry in bedrock at Leadhills-Wanlockhead are limited. The highest concentration of gold recorded in bedrock in the Leadhills-Wanlockhead area is from a felsic minor intrusion containing 413 ppb Au (Boast and Harris, 1984).

## 8 Structural controls of mineralisation

The NE-SW Caledenoid D<sub>1</sub> trend exerts a primary structural control on the location of gold mineralisation (Lusty et al., 2012). With the exception of Black Stockarton Moor and Glendinning, both located southeast of the OBF, the gold mineralised localities are spatially associated with major D<sub>1</sub> structures (Figure 1; Lusty et al., 2012). At the prospect scale, the auriferous veins and lodes are mostly ~N-S trending (e.g. Clontibret, Glendinning, Glenhead; Duller et al., 1997; Gallagher et al., 1983; Leake et al., 1981; Morris, 1984). However, in soil and deep overburden geochemical data (As, Au) from Glenhead, Hare Hill and Moorbrock Hill

the prominent trend is ~NE-SW and the ~N-S trend is subordinate (Beale, 1984; Boast et al., 1990; Leake et al., 1981). This contrasts with bedrock geochemical and structural data from drill core from Glenhead in which concordant ~NE-SW structures yield weak anomalies (<0.2 ppm Au; Figure 4), whereas discordant ~N-S faults and fractures give strong gold anomalies <8.8 ppm (Leake et al., 1981). At Hare Hill and Glenhead ~N-S anomalies correspond to zones of sericite alteration containing quartz veins ~1 cm thick (Boast et al., 1990; Leake et al., 1981).

Localised D<sub>2</sub> structural anomalies could have focussed synchronous and/or subsequent hydrothermal activity. In the area of Glenhead Burn (Figure 1) the regional strike swings to a more E-W orientation accommodated by near vertical folds in argillaceous rocks to the south (Leake et al., 1981; Stone and Leake, 1984) and is likely to have generated a deeply penetrating connected fracture system and enabled rapid ascent of hydrothermal fluids from depth. At Black Stockarton Moor the regional strike swings N-S forming a steeply-plunging sigmoidal monoform, possibly as a response to magmatic pressure from the Criffel-Dalbeattie Pluton (Brown et al., 1979). However, in other areas, e.g. Clontibret, D<sub>2</sub> structures are considered to be of little or no significance to gold mineralisation (Morris, 1984). Both D<sub>2</sub> and D<sub>3</sub> structures are likely to be related to the onset of regional transpression around 425 Ma (Miles et al., 2016).

At the deposit scale the lodes are most commonly controlled by transverse ~N-S to ~NW-SE trending (D<sub>3</sub>) structures (Figure 2, 3, 4), e.g. Clontibret, Glenhead Burn, Hare Hill, Fore Burn and Glendinning (Boast et al., 1990; Charley et al., 1989; Duller P.R, 1987; Duller et al., 1997; Leake et al., 1981; Steed and Morris, 1986). Abandoned mine plans from Leadhill-Wanlockhead indicate that the Pb-Zn mineralised structures trending ~N-S dip steeply to the east while those trending ~NW-SE dip steeply to the south-east. The lode zones at Clontibret are oriented ~140°/65°SW and are obliquely transected by ~N-S striking subvertical strike-slip faults containing muddy gouge up to several cm wide and fragments of mineralised and unmineralised rocks (Morris, 1984; Morris et al., 1986). Most of the Au-in-soil anomalies identified in central Ireland are elongate transverse to the dominant Caledonoid structural grain e.g. at Slieve Glah (anonymous, 2014b). Significant gold grades are found within ~N-S striking strike-slip faults within greywacke and intrusive igneous rocks at Glenhead (Figure 1, 3, 7; Leake et al., 1981; Stone and Leake, 1984). A N-S trending top-of-bedrock Au, As and Sb anomaly at Hare Hill corresponds to a ~N-S trending zone of subvertical to steeply west-

dipping veins within sericitised granodiorite containing disseminated arsenopyrite. The zone is cut by subparallel late-stage Sb-Pb veins (Boast et al., 1990). The Glendinning Sb deposit with auriferous arsenopyrite is controlled by a 015°-trending fracture system (Duller et al., 1997). The two ore zones at Fore Burn correspond to ~NW-SE-trending fault zones with irregular quartz-carbonate-sulphide veins, stockworks and breccia (Charley et al., 1989).

The ~N-S structural control is also expressed more cryptically. For example, the alluvial gold field at Leadhills-Wanlockhead lies at the intersection of a ~N-S trending topographic lineament that extends from the fault-controlled Carboniferous-Triassic Thornhill Basin, ~7km to the south with the D<sub>1</sub> ~NE-SW trending Leadhills Fault (Leake et al., 1998; Temple, 1956; Wilson and Flett, 1921) and the Palaeogene regional Eskdalemuir Dyke that exploits the ~NW-SE D<sub>3</sub> structural trend and could have remobilised gold (Leake et al., 1998; Macdonald et al., 2009). The magmatic PGE sulphide occurrence at Talnotry lies on a ~N-S trending line with the only other two such deposits known in Scotland at Sron Garbh in the Southern Highlands and Loch Ailsh in the Northern Highlands (Graham et al., 2017). In addition, locally developed late spaced cleavage with a strike of 110° corresponds to a topographic lineament set that intersects the reputed site of historic gold workings at Bulmers' Moss near Leadhills (pers. obs.; Gillanders, 1976; Porteous, 1876). The 110° trending topographic lineament also intersects Hare Hill and is weakly represented in soil As geochemical anomalies at Glenhead (Figure 4).

In summary, structural data indicate that at the regional scale the location of gold mineralisation is controlled by intersections between major NE-SW trending Caledonoid shear zones such as the Leadhills Fault and Slieve Glah Shear Zone with significant N-S and/or NW-SE trending transverse D<sub>3</sub> faults. At some localities, for example in Central Ireland near Slieve Glah, at Glenhead, Hare Hill and possibly Moorbrock Hill there is evidence at the prospect scale for ~NE-SW D<sub>1</sub> Caledonoid structural control of geochemical anomalies and gold grades. However, the predominant control of the orientation of mineralised lodes at the prospect scale appears to be the subvertical to steeply-dipping transverse D<sub>3</sub> faults and fractures trending either ~N-S or ~NW-SE.

## 9 Alteration

All of the gold-mineralised localities, whether hosted by, associated with or remote from known igneous intrusions exhibit similar phyllic to propylitic alteration assemblages. Auriferous veins at all of the localities exhibit envelopes of sericitised rock indicating that Au mineralisation was accompanied by peak hydrothermal potassic alteration (Figure 6). Other vein generations lacking alteration haloes may cut, or be cut by these veins, enabling recognition of pre-, syn- and post-alteration vein generations (Duller et al., 1997; Leake et al., 1981; Morris et al., 1986). Zones of alteration associated with disseminated or quartz vein-hosted auriferous sulphide mineralisation occur within metasedimentary and igneous host rocks and are <18 m wide (Boast et al., 1990; Steed and Morris, 1986).

Mineralogical zonation of potassic alteration assemblages characteristic of porphyry-type Cu-Au deposits is reported from Clontibret, Glendinning, Black Stockarton Moor and Glenhead Burn (Brown et al., 1979; Duller et al., 1997; Leake et al., 1981; Morris et al., 1986). However, although the same alteration mineral assemblages are preserved, porphyry type zonation appears to be absent at other localities e.g. Hare Hill and Fore Burn (Boast et al., 1990; Charley et al., 1989). Zoned phyllic-propylitic alteration is clearly related to intrusion of granodiorite sheets and the release of a magmatic-hydrothermal fluid at Black Stockarton Moor, where zoned alteration is developed within hydro-fractured metasedimentary rocks directly above porphyritic granodiorite sheets (Figure 6; Brown et al., 1979). Although established gold grades are very low at Black Stockarton Moor, very few Au determinations have been made. However, the relationships between alteration mineralogy and metasomatic enrichment of As, Sb and other pathfinder anomalies at Black Stockarton Moor are comparable to more richly gold-mineralised localities in the SUDLT (Brown et al., 1979; Duller et al., 1997; Leake et al., 1981; Shaw et al., 1995; Steed and Morris, 1986) and therefore indicate a common origin. Sericitic alteration at Black Stockarton Moor predominantly occurs within narrow zones around veins and above porphyritic granodiorite sheets (Brown et al., 1979). The sericitic zones are bleached and exhibit a pink colouration. The alteration assemblage comprises sericite, quartz, dolomite, muscovite and hematite developed pervasively with interstitial and replacive textures and filling veins (Brown et al., 1979). Orange colouration is locally associated with quartz-dolomite veins (Brown et al., 1979). The rocks in the sericitic zone contain abundant secondary pyrite, minor chalcopyrite and trace

bornite, molybdenite, tennantite and arsenopyrite (Brown et al., 1979). Chalcocite, enargite, covellite and sphalerite occur throughout (Brown et al., 1979). In addition, zones and patches of argillic alteration are developed locally in which kaolinite replaces plagioclase (Brown et al., 1979). The propylitic alteration assemblage in the metasedimentary country rocks at Black Stockarton Moor comprises chlorite, calcite, actinolite, epidote, albite, titanite and hematite plus jasperoid and minor sericite (Brown et al., 1979). Quartz-carbonate veinlets and metasomatic replacement veins are locally developed containing secondary actinolite, epidote and albite (Figure 6; Brown et al., 1979). In igneous rocks, the propylitic alteration assemblage consists of chlorite, replacing primary mafic silicates; hematite replacing primary oxides, and disseminated epidote and minor sericite replacing plagioclase feldspar (Brown et al., 1979). The propylitic alteration zones contain secondary disseminated pyrite and minor veinlets of chalcopyrite (Brown et al., 1979).

Discordant N-S trending subvertical veins with prominent phyllic alteration haloes at Glenhead exhibit zonation with an inner core composed of quartz, actinolite, diopside, magnetite, pyrrhotite, sericite, pyrite and sphene surrounded by an envelope rich in actinolite together with magnetite and ilmenite (Figure 8; Leake et al., 1981). Feldspars are sericitised and carbonated and mafic minerals are chloritised, sericitised and sulphidised. Actinolite from Glenhead Burn is comparable in composition to that in the propylitic alteration zone at Black Stockarton Moor, indicating similar conditions of hydrothermal alteration and mineralisation (Leake et al., 1981). At Fore Burn, metasomatic alteration affects intermediate to acidic volcanic and intrusive rock surrounding the lode zones are affected by intense potassic alteration with the assemblage sericite, chlorite, tourmaline, carbonate, quartz and apatite. The lode zones and individual auriferous fourth generation veins exhibit zonation of phyllic and propylitic alteration assemblages at Clontibret (Figure 6; Morris et al., 1986). In the outer, propylitic zone, secondary sericite is abundant in the matrix and replacing feldspathic detrital grains. Mafic detrital grains are completely replaced by oxychlorite and saussurite (Morris et al., 1986). Pale green chlorite occurs interstitially in patches and microcrystalline carbonate, together with chlorite forms overgrowths and veinlets (Morris et al., 1986). The inner (phyllic) zone contains more abundant sericite and carbonate. Veins of sericite, quartz and carbonate are accompanied by secondary arsenopyrite and pyrite (Morris et al., 1986). Interstitial chlorite is absent and secondary oxychlorite replaces mafic grains and is, in turn, partially replaced by sericite, indicating a progression of alteration from propylitic to sericitic and an



increase in  $a\text{H}_2\text{O}$  and addition of potassium (Morris et al., 1986). Detrital chromite grains exhibit haloes of bright green fuchsite (Cr-rich mica).

In summary, the relationship between hydrothermal alteration and gold mineralisation, in conjunction with other lines of evidence, helps to constrain the geochemical and physical conditions of mineralisation. It demonstrates a clear association with late Caledonian calc-alkaline magmatism. Comparisons with other mineral deposits globally and established mineral deposit models indicate that gold mineralisation in the SUDLT was related to magmatic-hydrothermal processes at shallow crustal levels,  $<\sim 8$  Km, comparable to the porphyry-epithermal spectrum of deposits (Berger et al., 2008; Brown et al., 1979; Richards, 2009; Richards et al., 2006; Steed and Morris, 1997). Furthermore, recognition of the association between gold mineralisation and peak potassic hydrothermal metasomatism is a useful aid to exploration for gold because it enables mineralised vein systems to be more easily identified and targeted.

## 10 Geochemistry

At all of the gold mineralised localities arsenic is sympathetically related to gold content and to indicators of potassic alteration (Figure 8; Boast et al., 1990; Duller et al., 1997; Leake et al., 1981; Morris et al., 1986). The association with arsenic reflects the abundance of auriferous arsenopyrite in which gold forms a lattice constituent (Duller P.R, 1987; Duller et al., 1997; Leake et al., 1981; Morris et al., 1986). Gold concentrations up to 3000 ppm have been recorded in arsenopyrite from Glenhead Burn and up to 2500 ppm in arsenopyrite from Clontibret (Leake et al., 1981; Morris et al., 1986). Gold is also present in pyrite. The auriferous arsenopyrite-pyrite assemblage is spatially associated with the most intensive wall rock alteration (Duller et al., 1997; Leake et al., 1981; Morris et al., 1986) and is reflected by a close correlation between As and the  $\text{K}_2\text{O}:\text{Na}_2\text{O}$  ratio, e.g. at Glendinning (Figure 8; Duller et al., 1997). The inner, phyllic, zone of hydrothermal mineralisation and alteration at Glendinning, Clontibret and Hare Hill corresponds to a relative enrichment of  $\text{SiO}_2$ ,  $\text{K}_2\text{O}$ ,  $\text{CaO}$ , S and As. The outer, propylitic zone is depleted in Na, Fe, Mg relative to the surrounding unaltered rocks (Boast et al., 1990; Duller et al., 1997; Morris et al., 1986). The most significant chemical expression of alteration is the increase in the  $\text{K}_2\text{O}:\text{Na}_2\text{O}$  ratio with proximity to the lode zones (Figure 8; Duller et al., 1997; Morris et al., 1986). The increase in  $\text{K}_2\text{O}$  reflects the abundance

of sericite. The inner zone of silicified and sericitised rock at Glendinning exhibits high CaO, SiO<sub>2</sub>, As, Sb and S values (Duller et al., 1997). High S (>25 000ppm) and As values correspond to zones of disseminated arsenopyrite and pyrite (Duller et al., 1997). The outer zone, up to 400 m wide is depleted in Na, Zn, Fe, Mn and Mg (Duller P.R, 1987; Duller et al., 1997). The outer zone corresponds to a Na<sub>2</sub>O depletion of up to 0.5%. K<sub>2</sub>O values reflect increased sericite abundance (Duller P.R, 1987; Duller et al., 1997). A sharp decrease in the MgO:CaO ratio marks the transition between the phyllic and propylitic zones (e.g. Clontibret, Figure 8) reflecting the addition of carbonate and the replacement of chlorite, feldspar and mafic minerals by sericite (Morris et al., 1986). Rubidium increases toward the lode in parallel with K<sub>2</sub>O reflecting the presence of Rb as a lattice constituent in micas (Morris et al., 1986). Calcium also increases while MgO and FeO decrease with proximity to the lode zone at Clontibret and Glendinning (Duller et al., 1997; Steed and Morris, 1986). Gold-bearing pyrite + arsenopyrite mineralised samples from Clontibret are relatively enriched in Bi, Ni, Co, Cu and Zn. Bi is a minor constituent of arsenopyrite, Co and Ni are lattice constituents in pyrite and Cu and Zn are present as inclusions of tetrahedrite, chalcopyrite and sphalerite within pyrite (Morris et al., 1986).

Increased CaO is associated with mineralisation at some localities, e.g. Clontibret and Black Stockarton Moor (Brown et al., 1979; Morris et al., 1986). However, at other localities, for example Glendinning and Hare Hill, an inverse relationship is seen between CaO and S (Boast et al., 1990; Duller et al., 1997). This could possibly have resulted from the dissolution of carbonate by acidic sulphidic mineralising fluids. Carbonate dissolution could have enhanced porosity and focussed subsequent episodes of mineralisation (Duller et al., 1997). Decreasing Mn, Fe and Zn levels with proximity to the mineralised zone accompanied by increase in Ca, for example, at Black Stockarton Moor is comparable to porphyry copper deposits e.g. the Kalamazoo deposit (Chaffee, 1975). In the Duns area, Cu does not correlate with enrichments of Au, Sb or As indicating that the Ba-Cu and Au-As-Sb mineralising events occurred separately (Shaw et al., 1995). the same relationships are seen in limited data for Au, As, Sb and Cu at Black Stockarton Moor (Brown et al., 1979).

Stibnite mineralisation is localised within the lodes at Clontibret, Glendinning and Hare Hill. Antimony mineralisation is not accompanied by wall rock alteration at Clontibret and exhibits a poor correlation with the K<sub>2</sub>O:Na<sub>2</sub>O ratio (Figure 8) indicating that stibnite mineralisation represents a separate event that may slightly post-date gold mineralisation

(Morris et al., 1986). That some correlation is evident reflects the fact that Sb mineralisation occupies the same, previously mineralised lode zones (Morris et al., 1986). Highly elevated Zn values at Glendinning <1846ppm occur within the mineralised fracture system due to sphalerite. However, away from the mineralised vein Zn is correlated with Pb and Sb but inversely with Ni, As and Co (Duller et al., 1997). This indicates the narrow zones of Zn enrichment reflect late-stage sphalerite mineralisation superimposed on a wider zone of Zn depletion resulting from the early-stage As-Sb-Au mineralisation (Duller et al., 1997). However, the relationships of some elements are not consistent everywhere. For example, Zn is depleted in the lode at Glenhead, Black Stockarton Moor and Glendinning relative to background levels, but enriched at Clontibret; Mn is depleted in the lode zone relative to background levels at Glendinning and Black Stockarton Moor, but is enriched at Glenhead; Ni and Rb are also enriched at Clontibret but depleted at Black Stockarton Moor. These geochemical differences could reflect local differences in the degree of alteration and temperature due to the exhumed level of the mineralised system or distance from the mineralised lode or differences in primary lithology of the host rocks. For example, the Portpatrick Formation contains a relatively high proportion of pyroxene and spinel (Oliver et al., 2003) that would contain Pb and Zn respectively. Detailed, integrated mapping, isotopic and petrological studies could resolve these possibilities. In summary, the geochemical data are consistent with the mineralogical and structural evidence that gold mineralisation was associated with the peak of late Caledonian magmatic-hydrothermal potassic hydrothermal alteration.

## 11 Fluid inclusions

Two distinct mineralising fluids are recognised in both auriferous veins and later Pb-Zn-bearing veins throughout the SUDLT (Figure 9,

Table 3). The relatively early, auriferous quartz veins contain rare inclusions of a 2-3 phase carbonic fluid with high homogenisation temperatures (158-386°C) and low salinity (0-11.7 wt.% NaCl) interpreted as a metamorphic and/or magmatic fluid (Baron and Parnell, 2000; Baron and Parnell, 2005; Moles and Nawaz, 1996).

Veins containing auriferous arsenopyrite-pyrite and stibnite mineralisation at Clontibret contain three-phase fluid inclusions composed of: aqueous liquid, CO<sub>2</sub> liquid and

CO<sub>2</sub> vapour. Salinity of the aqueous phase is estimated to be between 2 and 4 wt. % NaCl equivalent (Figure 9; Steed and Morris, 1986). Homogenisation temperatures are between 170°C and 340°C with a sharp peak at 290-300°C (Steed and Morris, 1986). The calculated fluid temperature is ~330°C based on an estimated minimum pressure of ~500 bars (Steed and Morris, 1986). At Glendinning fluid inclusions are rare in the early-stage arsenopyrite-quartz veins (Duller et al., 1997). Low-salinity (0-3 wt. % NaCl equiv.), CO<sub>2</sub>-rich, complex three-phase inclusions <5µm in diameter revealed temperatures in the range of 250-300°C and periods of boiling, comparable to those estimated for Clontibret (Figure 9; Duller et al., 1997). Primary fluid inclusions in early quartz veins from Leadhills-Wanlockhead have low salinities in the range 2 to 8 wt. % equivalent, some with very low salinities of 0 to 3.1 wt. % equivalent (Figure 9, 10). Fluid homogenisation temperatures for early quartz veins at Leadhills-Wanlockhead are between 187°C and 236°C (Figure 10; Samson and Banks, 1988). Primary inclusions from greywacke-hosted veins at Black Stockarton Moor contain liquid and vapour with a salinity between 4 and 12% NaCl and homogenisation temperatures between 197 and 386° (Figure 9; Lowry et al., 1997). Primary fluid inclusions from intrusive-hosted veins at Black Stockarton Moor exhibited some remarkably high salinities (up to 52 wt.% NaCl) and temperatures up to 468°C (Lowry et al., 1997). Fluid inclusion data for veins from Moorbrock Hill, Hare Hill and Stobshiel contain carbonic 2- and 3-phase inclusions containing H<sub>2</sub>O-CO<sub>2</sub>-CO<sub>2</sub> vapour or H<sub>2</sub>O+CO<sub>2</sub> vapour and exhibit low to moderate salinities in the range 0 to 9.24 equiv. wt% NaCl and have homogenisation temperatures between 190 and 250°C (Naden and Caulfield, 1989), consistent with the data for the early vein stage from the other localities (Figure 9). The early stage fluid is also identified in relatively early generations of veins at localities where gold has not been found, indicating that the gold-mineralising fluid flow event was widespread but generated only localised concentrations of gold. The fluid inclusion data are compatible with a magmatic-hydrothermal origin for gold mineralisation (Duller et al., 1997; Lowry et al., 1997; Naden and Caulfield, 1989; Steed and Morris, 1997) and are also consistent with the pressures and temperatures indicated by the low grade prehnite-pumpellyite metamorphic mineral assemblage in the metasedimentary rocks (Oliver, 1978).

A later, low temperature (5-228°C) higher salinity (1.4-29 wt.% NaCl) aqueous fluid of meteoric origin is associated with the Pb-Zn mineralisation and has been interpreted as being of Carboniferous age (Baron and Parnell, 2000; Baron and Parnell, 2005; Ineson and Mitchell, 1974; Lowry et al., 1997; Moles and Nawaz, 1996; Samson and Banks, 1988). The relatively

late, Pb-Zn sulphide veins throughout the SUDLT contain fluid inclusions with generally higher salinities and lower homogenisation temperatures (Table 3; Figure 9, 10; Baron and Parnell, 2005; Samson and Banks, 1988). Late stage quartz and dolomite from Conlig-Whitespots and Castleward (Figure 1) contains two-phase H<sub>2</sub>O-salt inclusions with salinities between 1.4 and 15.86 wt% NaCl equiv. and homogenisation temperatures 83°C to 228°C (Baron and Parnell, 2005). Inclusions in late-stage veins from Leadhills-Wanlockhead have lower homogenisation temperatures that range from 5 to 134°C (Samson and Banks, 1988). Secondary inclusions from Black Stockarton Moor are CO<sub>2</sub>-deficient, H<sub>2</sub>O dominated, homogenise at 123-188°C and have salinities up to 22 wt% equiv. NaCl (Lowry et al., 1997). The late stage fluid is interpreted as a low temperature meteoric basinal brine and must represent markedly different conditions and a different mineralising episode from the earlier higher temperature gold event, and therefore, is interpreted as significantly younger (Figure 9, 10).

The data clearly reveal two distinct fluid types with distinct chemical compositions and representing markedly different physical conditions and, therefore, two distinct episodes of hydrothermal mineralisation (Figures 9, 10; Baron and Parnell, 2005; Samson and Banks, 1988). these two fluids are recorded in gold-mineralised rocks within or demonstrably genetically associated with late Caledonian igneous rocks and within metasedimentary host rocks remote from any known igneous intrusions. Comparable distinct early and late mineralising fluids have also been identified in the Dalradian rocks of the Grampian Highlands Terrane (Baron and Parnell, 2000; Treagus et al., 1999; Wilkinson et al., 1999). In addition, Lowry et al. (1997) documented very high salinity (<52 wt.% NaCl) fluid inclusions with high homogenisation temperatures (<532°C) from quartz veins within intrusive igneous rocks at Black Stockarton Moor and Cairngarroch Bay. These fluids must represent coeval Caledonian magmatic-hydrothermal mineralising fluids and it seems likely that this fluid migrating along faults and fractures to form satellite deposits.

## 12 Sulphur Isotopes

Hydrothermal sulphide associated with Caledonian gold concentrations exhibits  $\delta^{34}\text{S}$  values in the range -4.9 to +6 ‰. This range is significantly higher than, but overlaps with the  $\delta^{34}\text{S}$  values for diagenetic sulphide in the Moffat Shale from Clontibret and Leadhills that range between -0.6 to -17.1 ‰ (mean = -8.4 ‰; Figure 11; Anderson et al., 1989). Two

samples of pyrite from unmineralised shale at Clontibret have  $\delta^{34}\text{S}$  values of -15.1 and -15.7 ‰ (Figure 11; Anderson et al., 1989). The difference in these two ranges could be most easily explained if some of the samples of Moffat Shale from Leadhills were hydrothermally mineralised. This is considered likely, as pervasive deformation, veining and sulphide mineralisation are observed in the Moffat Shale in the Leadhills area (pers. obs.; Temple, 1956; Wilson and Flett, 1921), where it forms the decollement of the Leadhills Imbricate Zone and is, therefore, likely to have been the locus of hydrothermal fluid flow. This is supported by the observation that at Clontibret  $\delta^{34}\text{S}$  of hydrothermal sulphide within the lodes is consistently greater than it is for diagenetic pyrite in the unmineralised Moffat Shale (Figure 11; Steed and Morris, 1997).

$\delta^{34}\text{S}$  values for sulphide minerals from a gold-bearing vein within the Fleet pluton near Talnotry (Orchars Vein; Figure 1) are between -12 and -5 ‰; comparable with those for Pb-Zn veins at Leadhills-Wanlockhead (Anderson et al., 1989), and therefore are considered to reflect low temperature meteoric remobilisation during a significantly later episode of structural reactivation (Anderson et al., 1989; Baron and Parnell, 2000; Lowry et al., 1997; Lusty et al., 2011; Samson and Banks, 1988).

Excluding the Orchars Vein,  $\delta^{34}\text{S}$  values for sulphides in veins and wallrocks from gold-bearing localities in the SUDLT range between -4.9 and + 6 ‰ (Figure 11). All of the available S isotope data are from gold-bearing localities that are spatially associated with known intrusions, with the exception of Glendinning and Clontibret. However, metasedimentary rocks are hornfelsed in the vicinity of Clontibret and minor Caledonian intrusions do occur. Glendinning is within the area of the possible buried Tweedale pluton (Stone et al., 2012). Reported  $\delta^{34}\text{S}$  values for Glendinning and Clontibret taken together range between -3.95 and +6.0 ‰ (Figure 12; Duller et al., 1997; Steed and Morris, 1997) whereas  $\delta^{34}\text{S}$  values for auriferous lodes spatially associated with igneous intrusions fall in the slightly lower, but significantly overlapping range between -4.9 to +2.8 ‰ (Figure 11; Lowry et al., 1997; Naden and Caulfield, 1989). The narrow range of sulphide  $\delta^{34}\text{S}$  from Glendinning, remote from any known intrusion, is very similar to that for intrusion-related mineralisation at Talnotry, Cairngarroch and Hare Hill, but also overlaps with the range of values for Clontibret. However, Clontibret exhibits a greater range of  $\delta^{34}\text{S}$  that extends to higher values. There are insufficient S isotope data from localities demonstrably remote from any known Caledonian igneous intrusions to make any inference about the role of sedimentary versus magmatic sulphur.

However, the overall difference between the ranges for diagenetic and hydrothermal sulphides indicates an external sulphur input, possibly of magmatic origin. Excepting Black Stockarton Moor and Clontibret, the  $\delta^{34}\text{S}$  values for hydrothermal pyrite from the gold-bearing localities fall within the upper part of the range for diagenetic pyrite in the Moffat shale (Anderson et al., 1989). The narrow range of  $\delta^{34}\text{S}$  values exhibited by these localities indicate input of magmatic S, either directly or by subsequent leaching of igneous rocks (Duller et al., 1997). This is supported by the observation that massive replacement sulphide in a diorite intrusion at Cairngarroch Bay exhibits more enriched  $\delta^{34}\text{S}$  (mean -1.9‰) than arsenopyrite from associated quartz veins within the metawacke country rocks (mean -2.8‰; Lowry et al., 1997). However, the overlapping S isotope data indicate that sulphides in the country rocks may have dissolved in the circulating hydrothermal fluids (Lowry et al., 1997). The dissolution of sulphide in the fluid would have contributed to the capacity for the hydrothermal fluid to carry and transport gold-sulphide complexes in greater concentrations and over greater distances, enhancing the capacity for economic gold concentrations regionally.

### 13 Oxygen and hydrogen isotopes

Silicate minerals in the hydrothermally altered and mineralised zones exhibit higher  $\delta^{18}\text{O}$  values than the unmineralised rocks (Naden and Caulfield, 1989). Measured  $\delta\text{D}$  and  $\delta^{18}\text{O}$  values for minerals and fluid inclusions in early (Caledonian gold phase) and late (Pb-Zn-Cu) veins are shown in Figure 12, together with calculated  $\delta^{18}\text{O}$  values for the mineralising fluids.  $\delta^{18}\text{O}$  was not reported for the early quartz veins at Leadhills. The quartz vein at Cairngarroch Bay has  $\delta^{18}\text{O}$  of +11.6‰ and contains fluid inclusions with  $\delta\text{D}$  of -46‰. Vein quartz at Talnotry has  $\delta^{18}\text{O}$  of +14‰ with fluid inclusions with a  $\delta\text{D}$  of -53‰ (Lowry et al., 1997). The calculated  $\delta^{18}\text{O}$  for the mineralising fluid is +8.4‰ for Cairngarroch Bay and +9.6‰ for Talnotry (Lowry et al., 1997). Samples of quartz from auriferous arsenopyrite veins at Clontibret (for which there are no  $\delta\text{D}$  data) yielded  $\delta^{18}\text{O}$  values between +14.6 to +19.2 with a mean of +17.0‰ (SMOW; Steed and Morris, 1997). Sericite from altered greywacke and felsic igneous rocks from the lode zone at Clontibret has  $\delta^{18}\text{O}$  between +12.2‰ to +14.3‰ with a mean of +13.2‰ (Steed and Morris, 1997). The  $\delta^{18}\text{O}$  of the mineralising fluid was calculated to be

926 +10.5 ‰ (SMOW) and  $\delta D$  -30‰ using the fluid temperature of 330°C (Steed and Morris,  
927 1997).

928 The  $\delta^{18}O$  and  $\delta D$  values for quartz veins at Cairngarroch Bay and Talnotry fall clearly  
929 within the range for magmatic fluids and overlap with the field for metamorphic fluids (Lowry  
930 et al., 1997). The values from Clontibret correspond fairly well with those for Talnotry and  
931 Cairngarroch Bay, but the calculated isotopic composition of the fluid falls outside the range  
932 for magmatic water. However, fluid isotope values should not be considered to directly reflect  
933 the isotopic composition of the primary source fluid because fluid rock interaction greatly  
934 influences fluid isotopic compositions (Boehlke and Kistler, 1986). The narrow range of  $\delta^{18}O$   
935 values for Clontibret, Talnotry and Cairngarroch Bay indicates that the mineralising fluid is  
936 unlikely to have been an evolved meteoric fluid because fluid-rock interaction is not likely to  
937 produce such a narrow isotopic range (Steed and Morris, 1997).

938 Late base metal veins in the Southern Uplands have  $\delta D$  values between -40 and -70‰  
939 and  $\delta^{18}O$  values that range from -7.5 to +6.5 ‰ (Figure 12; Samson and Banks, 1988). The  
940  $\delta^{18}O$  value of one of these samples is so low that it indicates a fluid  $\delta^{18}O$  below the lower limit  
941 for meteoric water and is therefore unreliable. The measured  $\delta D$  constrains the minimum  
942 possible  $\delta^{18}O$  value for the fluid to -7.5 ‰. These data indicate a minimum precipitation  
943 temperature of ~110°C, which is consistent with the measured fluid inclusion homogenisation  
944 temperatures (Samson and Banks, 1988). On a regional scale, the range of  $\delta D$  and  $\delta^{18}O$  values  
945 for the Southern Uplands Pb-Zn veins predominantly lie between the compositions of  
946 meteoric water and magmatic and metamorphic fluids (Samson and Banks, 1988). The very  
947 low calculated fluid  $\delta D$  for some of the Pb-Zn veins suggests that metamorphic fluids were  
948 not involved in the Pb-Zn mineralisation (Samson and Banks, 1988). This is consistent with the  
949 marked differences between the Pb-Zn veins and the Caledonian (Au) quartz veins in terms  
950 of fluid inclusion compositions and homogenisation temperatures mineral assemblage  
951 paragenesis that indicate that the Pb-Zn-Cu veins significantly post-date Caledonian  
952 orogenesis. Low fluid temperatures and a lack of consistent spatial correlation with igneous  
953 intrusions indicates that the late stage fluid was of purely meteoric origin, isotopically  
954 modified by interaction with igneous and metamorphic rocks (Samson and Banks, 1988).  
955



## 14 Discussion

It has long been recognised that orogenic belts host a wide range of gold deposit types and that there are many overlapping characteristics between orogenic gold deposits, intrusion related gold systems (IRGS) and postsubduction porphyry gold (Groves et al., 1998; Hart, 2005; Richards, 2009). Groves et al. (1998) raised the question of whether or not IRGS should be included in the orogenic class of gold deposit types, which overlap in terms of post peak-metamorphic timing, low to moderate salinity H<sub>2</sub>O-CO<sub>2</sub> fluids, 3-20 km depths of ore deposition, low concentrations of base metals (Cu, Pb, Zn), significant additions of As, Bi, Sb, Te and W and hydrothermal gains of K, S and Si (Goldfarb et al., 2001; Groves et al., 1998; Sillitoe, 1991; Sillitoe and Thompson, 1998; Thompson et al., 1999). Many orogenic gold deposits have been reclassified as IRGS due to their shared characteristics and common spatial association with intrusions in orogenic settings (Groves et al., 1998; Hart, 2005). The term reduced intrusion-related gold system (RIRGS) was subsequently introduced to distinguish more clearly between this deposit type and orogenic gold, leaving oxidised IRGSs to be classed as either orogenic or porphyry type (Hart, 2005). However, such definitions are restrictive and the deposit model approach has limited ability to adequately describe the range of mineral deposits that may be transitional between idealised types. Debate about classification is further complicated by continued debate about the sources of Au, S and fluid in orogenic deposits and whether or not magma is important for transporting Au, S and fluid from the lithospheric mantle and/or lower crust to shallower depths (Groves et al., 2003; Phillips and Powell, 2009). Furthermore, growing recognition of the role of transient geodynamic scenarios in mineralising systems indicates the potential for overlap between orogenic, IRGS and the continuum of porphyry, skarn and epithermal vein gold deposit types (Groves et al., 1998; McCuaig and Hronsky, 2014; Richards, 2009). For example, changing plate-boundary kinematics, postsubduction slab break-off and sub-continental lithospheric mantle delamination may cause pulses of anomalous magmatism, heat and hydrothermal activity at shallow crustal levels, in particular, in soft collisional settings, that can facilitate the mass and energy flux required for mineralisation (McCuaig and Hronsky, 2014; Richards, 2009). Groves et al. (1998) suggested that orogenic gold deposits cannot form at less than ~2.5 km depth due to gold solubility relationships below ~200°C. The recognition of postsubduction porphyry and epithermal vein gold deposits in orogenic settings (e.g.

Richards, 2009) indicates an important role for magmatic activity in orogenic settings for extending the range of ore-forming environments. This logic can be extended to IRGS, the widespread occurrence of which indicates that gold at shallow crustal levels in orogenic settings could be more common than previously thought. Slab break-off and/or delamination is a possible mechanism for generating IRGS, porphyry, skarn and epithermal type gold deposits in orogenic settings (Richards, 2009). In an alternative approach to exploring for a wide range of mineral deposit types at the regional scale, McCuaig and Hronsky (2014) advocate four critical elements of the general 'mineral system': lithospheric architecture, transient favourable geodynamics, fertility and preservation. Irrespective of the fit to the range of deposit models, data from the SUDLT indicate a good match with these criteria.

The timing of gold mineralisation in the SUDLT limits the possible models of mineralisation according to the well-constrained tectonic evolution of the terrane. Gold mineralisation is hosted by D<sub>3</sub> transverse faults and auriferous pyrite and arsenopyrite were the first sulphides to precipitate during initial brecciation and hydrothermal alteration (Duller et al., 1997). The age of mineralisation is therefore constrained by the maximum age of D<sub>3</sub> transverse faults, constrained by broadly contemporaneous lamprophyric dykes dated between 418 and 395 Ma (MacDonald et al., 1985; Rock et al., 1986). Minor intrusions and altered rocks are both hornfelsed in the contact metamorphic aureoles of the Criffel and Loch Doon plutons and are truncated by, and found as xenoliths within the plutonic complexes, indicating that dyke emplacement and hydrothermal mineralisation preceded emplacement of the large plutons (Brown et al., 1979). However, higher grade auriferous ~N-S veins cut the dioritic margin of the Loch Doon Plutonic Complex (Figure 7; Leake et al., 1981). These relationships indicate that gold mineralisation was polyphase and coeval with progressive emplacement of the Loch Doon Plutonic Complex at  $408 \pm 2$  Ma (Rb-Sr mineral-whole-rock age; Halliday et al., 1980). Brecciation and intense hydraulic fracturing are developed in altered metasedimentary rocks immediately above granodiorite sheets, indicating syn-magmatic hydrothermal alteration and mineralisation (e.g. Black Stockarton Moor; Brown et al., 1979). The granodiorite sheets are cut by the  $410 \pm 6$  Ma Criffel Pluton (zircon U/Pb age). Gold mineralisation therefore, most probably occurred between ~418 and ~410 Ma, i.e. closely following the final closure of Iapetus along the Solway Line (Dewey and Strachan, 2003; Miles et al., 2016). The latest Silurian to Early Devonian age indicates that mineralisation occurred following the arrival of the Avalonian continental margin at the

subduction trench during initial soft collision and the onset of regional transtension (Dewey and Strachan, 2003; Miles et al., 2016). Mineralisation occurred during a period of transient geodynamics accompanied by orogenic magmatism, transtensional deformation and delamination of the Avalonian sub-continental lithospheric mantle following the arrival of the Avalonian margin at the subduction trench (Freeman et al., 1988; Miles et al., 2016).

The very low grade metasedimentary rocks of the SUDLT range from late diagenetic to epizone facies, indicating maximum temperatures of around 300°C (Merriman and Roberts, 2000). The metamorphic map of Merriman and Roberts (2000; Figure 13) shows that metamorphic grade is not clearly related to stratigraphic age or structural position, indicating that metamorphism post-dates thrust imbrication of the subduction-accretion complex. The spatial pattern of metamorphic grades in SW Scotland appears to reflect two controls 1) contact metamorphism around plutons and 2) major strike-slip shear zones, for example the Moniaive Shear Zone. Hydrothermal fluids are the most effective means of heat transfer in the crust and are an important control of low grade metamorphism at shallow crustal levels (Jamtveit and Austrheim, 2010; Robb, 2009). The pattern of metamorphism in the SUDLT (Figure 13) most probably reflects the activity of magmatic-hydrothermal fluids in both transferring heat to shallow levels and lowering the temperatures of metamorphic reactions by increasing the  $a_{H_2O}$  (Jamtveit and Austrheim, 2010). These prograde metamorphic reactions would, in turn, have produced additional fluids and possibly released S and metals. Metamorphic grade contrasts across Caledonoid structures could, therefore, be explained by fault reactivation or permeability contrasts. The metamorphic map indicates that Caledonian intrusions and shear zones focussed low grade metamorphism and, most probably, coeval hydrothermal activity and mineralisation in the SUDLT. Furthermore, the distribution of arsenic reflects the pattern of low grade metamorphism (Figure 13) suggesting a genetic link with mineralization. The temperature range, timing and structural controls of metamorphism are compatible with a shallow orogenic-type gold mineralising system (Groves et al., 1998) and the metamorphic map supports syn-kinematic hydrothermal metamorphism related to magmatism, possibly during postsubduction recovery of a perturbed geothermal gradient or asthenospheric upwelling (Miles et al., 2016).

Gold mineralisation in the SUDLT was accompanied by peak phyllic-propylitic hydrothermal alteration (Allen et al., 1982; Boast et al., 1990; Brown et al., 1979; Duller et al., 1997; Leake et al., 1981; Shaw et al., 1995; Stanley et al., 1987; Steed and Morris, 1997).

Zonation of the alteration assemblages, comparable to porphyry-type deposits is common (Allen et al., 1982; Brown et al., 1979; Duller et al., 1997; Morris et al., 1986; Steed and Morris, 1997) but noted to be lacking at some localities (Boast et al., 1990), possibly due to subsequent deformation. Hydrothermal alteration and gold-arsenopyrite mineralisation are clearly related to late Caledonian intrusions at Black Stockarton Moor, Glenhead Burn, Fore Burn and Hare Hill.

Hydrofracturing, alteration and mineralization within immediate country rocks to minor intrusions at Glenhead Burn and Black Stockarton Moor demonstrate that mineralisation was directly related to shallow-level emplacement of granodiorites and monzonites. However, gold lodes with the same paragenesis, alteration mineralogy and geochemistry also occur apparently remote from any significant exposed intrusion, for example, Central Ireland, Glendinning and Leadhills (Boast and Harris, 1984; Duller et al., 1997; Morris et al., 1986; Figure 1). On the basis of isotope, fluid inclusion and mineralogical evidence Lowry et al. (1997) consider that the plutons were the source of heat but that fluids, sulphur and metals were derived from both the intrusions and the metasedimentary country rocks through hydrothermal processes. This suggests the very low metamorphic grade of the metasedimentary rocks at the time of late Caledonian magmatism was an important factor controlling the 'fertility' of the terrane with respect to mineralising fluids (Lowry et al., 1997). Steed and Morris (1997) suggested that the spatial association could be explained by the enhanced capacity for brittle fracturing and vein development resulting from contact metamorphism. Furthermore, the apparent spatial association between mineralisation and large intrusive complexes may simply reflect the shared geodynamic setting and structural controls, rather than any direct genetic association (Goldfarb et al., 2005; Tomkins, 2013). However, fracturing, metasomatism and mineralisation preceded final pluton emplacement at Black Stockarton Moor and Glenhead (Leake et al., 1981). Gold is spatially associated with the earliest more mafic parts of the plutons, e.g. at Glenhead for the Loch Doon pluton (Leake et al., 1981), Slieve Croob for the Newry Igneous Complex (Smith et al., 1996; Young, 1987) and Moorbrock Hill for the Cairnsmore of Carsphairn pluton (Beale, 1984; Dawson et al., 1977). In addition, the PGE-enriched magmatic sulphide cumulate deposit at Talnотry is a clear example of a magmatic source of gold, demonstrating that late Caledonian magmas were capable of transporting and concentrating precious metals (Power et al., 2004). The metal enrichment of the magma could have resulted from either crustal contamination or a

magmatic source metasomatically enriched in Au and S (Lowry et al., 1997). The lack of any consistent mineralogical, geochemical or structural differences between these and gold localities remote from known intrusions indicates a common process of gold mineralisation.

Sulphur isotope values provide further support for a fundamental link between gold deposition and magmatism (Duller et al., 1997; Lowry et al., 1997), as is the case for many orogenic and/or IRGS gold deposits globally e.g. Mother Lode deposit, USA (Steed and Morris, 1997), Wasamac and other deposits in the Abitibi Greenstone Belt, Canada (Meriaud and Jebrak, 2017). The  $\delta^{34}\text{S}$  data indicate that magmatic sulphur mixed with sedimentary sources (Naden and Caulfield, 1989). However, there are currently no data to suggest that gold mineralisation is spatially associated with the Caledonian lamprophyres that crop-out predominantly within the Southern Belt of the Southern Uplands. The lamprophyres may, however, represent the primary magma from which the other igneous rocks were derived. Therefore, analysis of their Au and S content could reveal whether or not the source region was enriched in Au and S.  $\delta^{34}\text{S}$  values -1 to +3 ‰ from intrusive-hosted sulphides at Black Stockarton Moor indicate a strong contribution of I-type magmatic sulphur within the range for magmas with a subcrustal source (Lowry et al., 1997). Subcrustal I-type magmatic sulphur was also the predominant source at Cairngarroch Bay and Talnotry with a greater contribution of sedimentary sulphur (<50%) evident in the greywacke-hosted veins (Lowry et al., 1997). A minor component of sedimentary sulphur is possible for the granitoid-hosted hydrothermal mineralisation for which  $\delta^{34}\text{S}$  values are mostly in the range -3 to 0 ‰, (Lowry et al., 1997). The range of  $\delta^{34}\text{S}$  values for hydrothermal mineralisation falls between subcrustal magmatic sulphur and the strongly depleted sulphur of the SUDLT metasedimentary rocks (Moffat Shale) indicating hydrothermal equilibration between fluids and host rocks and/or mixing between magmatic-hydrothermal fluid and fluid derived by contact metamorphic dewatering of the country rocks (Lowry, 1991; Lowry et al., 1997; Lowry et al., 2005). There is no evidence for deep burial, melting, assimilation or contamination of magma by metasedimentary rocks of the Southern Uplands. Indeed, Hf, O and Pb zircon isotopes provide convincing evidence for a magmatic source in underplated Avalonian crustal rocks comparable to the Skiddaw Slate of the Lakesman Terrane with no involvement of Southern Uplands material (Miles et al., 2014; Miles et al., 2016; Thirlwall, 1989). The association of gold mineralisation with the relatively early phase of more mafic, reduced I-type magmatism with a subcrustal S isotope signature indicates that the mantle source was relatively oxidised. It is well known that

oxidised magmas suppress early sulphide saturation and, therefore, have greater potential to transport gold-sulphide complexes for longer and over greater vertical crustal distances (Ishihara, 1981; Robb, 2009). Melting and assimilation of sulphide-rich crustal metasedimentary rocks, e.g. Skiddaw Group, would be expected to lead to more reducing magma compositions and possible over-saturation with respect to sulphide, thus removing gold-sulphide complexes as a dense immiscible melt. This is reflected by the lack of gold associated with the relatively late more silicic S-type granitoid inner zones of the TSS plutons with more reduced S-type compositions.

Two separate mineralising events are recognised that record distinctly different physical and chemical conditions, and therefore occurred at different times. The early phase was associated with Au-As-Sb and the later phase probably represents the regional Pb-Zn-Ag mineralising event of uncertain age. Both phases of mineralisation are evident within individual lodes indicating a common regional structural and metallogenic history of reactivation (Baron and Parnell, 2005). The moderate to high temperatures of the early inclusions are compatible with a late Caledonian origin (Baron and Parnell, 2005; Samson and Banks, 1988). The timing of the later, low temperature fluid is less certain and could have occurred at any time after the Caledonian. Two distinct fluid types, that must be of significantly different age, have also been recognised in Pb-Zn and Au deposits in the Dalradian rocks of the Grampian Highlands Terrane: Tyndrum in Scotland, for which a magmatic origin is favoured and Curraghinalt, Northern Ireland (Baron and Parnell, 2005; Craw and Chamberlain, 1996; Curtis et al., 1993; Rice et al., 2016; Wilkinson et al., 1999). Lowry et al. (1997) proposed that the secondary fluid represents meteoric water that was heated by the intrusions and mixed with magmatic fluids with precipitation caused by mixing between the two fluids. However, this is difficult to reconcile with the two consistently distinct fluid phases found in the veins regionally.

The early stage hydrothermal system initiated at 3-5 km depth within rocks of very low metamorphic grade and had low to moderate salinity and high CO<sub>2</sub> contents indicating at least a partially magmatic source (Duller et al., 1997; Lowry et al., 1997; Naden and Caulfield, 1989; Stone et al., 1995). Fluid inclusion data do not preclude a metamorphic origin for the early fluid. However, most of the veins appear to have formed from a mixture of magmatic and formation waters and contain sulphur of mixed origin. Some of the higher boiling temperatures of fluid inclusions from veins in the Southern Uplands within igneous host rocks

are comparable to those for porphyry gold systems (Lowry et al., 1997; Naden and Caulfield, 1989). The calculated  $\delta^{34}\text{S}$  for  $\text{H}_2\text{S}$  in the ore-forming fluid at Clontibret (about  $\pm 1\text{‰}$ ) is typical of orogenic gold deposits globally and is also compatible with derivation from an igneous source (Steed and Morris, 1997). However,  $\delta^{18}\text{O}$  values calculated for the primary ore fluid (e.g.  $+10.7\text{‰}$  at Clontibret) are slightly above the range for magmatic waters ( $+5.5$  to  $+10.0\text{‰}$ ; Taylor, 1979).  $\delta^{13}\text{C}$  values for dolomite at Clontibret indicate that the host rocks were not the dominant source of carbon for the ore fluid (Steed and Morris, 1997).

Fluid inclusions in late-stage veins within intrusions and country rocks are deficient in  $\text{CO}_2$ , and have higher salinity (5-14 wt.% NaCl equiv.) and lower homogenisation temperatures ( $<300^\circ\text{C}$ ) than fluid inclusions in the early veins (Figure 9; Baron and Parnell, 2005; Duller et al., 1997; Lowry et al., 1997; Samson and Banks, 1988; Steed and Morris, 1986; Steed and Morris, 1997). These characteristics indicate that the later fluid is unlikely to have been derived from either the intrusions or contact metamorphic dewatering of the country rocks but instead represents modified meteoric water or basinal brine (Lowry et al., 1997; Samson and Banks, 1988; Steed and Morris, 1986). The Pb-Zn deposits at Leadhills-Wanlockhead, Conlig-Whitespots and Castleward are hosted by carbonate veins that contain inclusions of a comparable aqueous fluid (Baron and Parnell, 2005; Samson and Banks, 1988). Sulphur isotope values indicate that sulphides from Pb-Zn veins at Wanlockhead represent leached and homogenised Lower Palaeozoic diagenetic sulphide (Anderson et al., 1989). The  $\delta^{34}\text{S}$  composition of the veins at Leadhills-Wanlockhead is close to the composition of pyrite in the Moffat Shale (Anderson et al., 1989). However, at some other Pb-Zn deposits in the region, e.g. Navan, much heavier  $\delta^{34}\text{S}$  indicates a contribution from deep-seated magmatic or metamorphic sulphur (Anderson et al., 1989). Carboniferous sedimentary Pb-Zn mineralisation in central Ireland (e.g. Navan) is considered likely to represent the same hydrothermal event as Pb-Zn mineralisation at Leadhills-Wanlockhead, Whitespots-Conlig and elsewhere in the SUDLT (Ineson and Mitchell, 1974; Temple, 1956).

Figure 14 shows our preferred model for the geodynamic controls of Caledonian gold mineralisation in the SUDLT. The mantle beneath the region is metasomatically hydrated and 'fertilised' by the history of northwards subduction Iapetus lithosphere. Soft collision results in slab delamination because the relatively slow advance of the down-going plate does not exceed the rate of vertical descent due to negative buoyancy of the subducted slab. Slab break-off and/or delamination of the downgoing sub-continental lithospheric mantle leads to

asthenospheric upwelling and generation of hot lamprophyric melt which migrates and conducts heat to the underplated Avalonian crustal rocks beneath the SUDLT causing melting and assimilation and generating calc-alkaline magmas. Following the final subduction of Iapetus oceanic lithosphere and the arrival of the continental margin at the subduction trench there is a transition from orthogonal convergence to transtension and strike-slip deformation. This gives rise to dilatant vertical structural favourable for the effective transfer of heat energy and mass to upper crustal levels. The crustal rocks of the Laurentian margin, including the SUDLT, were not deeply buried or highly metamorphosed during soft collision leaving them 'fertile' with respect to H<sub>2</sub>O and leading to low amounts of exhumation thus increasing the preservation potential of the gold mineralised system.

Magma is particularly important in these systems in conveying energy and mass to shallow crustal levels at low pressures and temperatures. Heat flow, magmatism and transient tectonics related to slab break-off and/or delamination are likely to have been important factors in conveying heat, fluids and metals to shallow crustal levels. In addition, Ordovician-Silurian subduction of Iapetus oceanic crust is likely to have metasomatically hydrated and fertilised the Laurentian lithospheric mantle with respect to sulphide and gold. Oblique soft collision between Avalonia and Laurentia in Wenlock time initiated deep subvertical strike-slip faults, representing a favourable crustal architecture for effective vertical mass transfer. Slab break-off and/or delamination of the Avalonian plate at ~420-405 Ma (Miles et al., 2016), thermal relaxation and coeval post-collisional regional transtension provided a favourable transient geodynamic scenario and an anomalously high heat flow necessary for economic gold mineralisation. The lack of crustal thickening inherent and the passive post-orogenic history has favoured the preservation of gold deposits. The processes of Caledonian gold mineralisation in the SUDLT explain the overlapping characteristics and possible continuum between orogenic, IRGS and post-subduction porphyry deposit types at shallow levels in soft-collisional settings.

The nature of gold mineralisation in the SUDLT demonstrates that within the context of soft continental collision and orogenesis magma can play an important role in the transfer of significant energy and mass. In rocks of very low regional metamorphic grade this can lead to overlapping processes and products (characteristics) between deposit models associated with different tectonic settings. For example, the mineralisation is strongly structurally controlled and syn-kinematic as in orogenic lode gold and is associated with magmatic-



hydrothermal potassic alteration as in porphyry Cu (Au) deposits in supra-subduction zone magmatic arcs. This indicates the limited capacity of traditional genetic deposit models to identify new deposit types and supports a role for the mineral systems approach (McCuaig and Hronsky, 2014).

## 15 Conclusions

This synthesis represents the first regional scale review of gold mineralisation in the SUDLT. The findings demonstrate that gold mineralisation is broadly spatially and temporally associated with the pattern of peak low grade metamorphism, phyllic-propylitic hydrothermal alteration and minor intrusions and is controlled, at the deposit scale by discordant, steeply-dipping transverse  $D_3$  structures. Mineralisation occurred in sub-greenschist facies conditions in the upper crust probably at <5 km depth and at temperatures between 300 and 400°C, consistent with conditions in the upper crust during Caledonian soft continental collision.

Cross-cutting relationships between mineralisation, related structures and dated lamprophyric and calc-alkaline intrusions indicate a latest Silurian to Early Devonian age (~418 and ~410 Ma) for gold mineralisation, and was therefore coeval with soft collision, regional transtension and slab break-off and/or lithospheric delamination (Dewey and Strachan, 2003; Miles et al., 2016; Stone, 1995). This scenario is supported by tectonic discrimination of lamprophyric minor intrusions that indicate a postsubduction geodynamic setting. The SUDLT is an accretionary complex within a Phanerozoic soft collisional orogen and is representative of Phanerozoic orogenic gold belts globally, e.g. Central Asian Tethysides. The range of deposit types within the SUDLT includes magmatic sulphide, intrusion-related, porphyry type and structurally hosted lode gold. These various types exhibit remarkably similar character in terms of mineralogy, fluid inclusion properties, conditions of formation and age, indicating a common origin. Together with the observations from some gold mineralised localities that exhibit a clear genetic relationship to magmatism this indicates that all Caledonian gold mineralisation in the SUDLT is likely to be related to magmatism. The magma was sourced in underplated Avalonian crust and mantle (Miles et al., 2014; Thirlwall, 1989). Oxidised I-type sub-crustal melts transferred gold to shallow crustal levels in the overriding Laurentian plate (e.g. at Talnotry and Black Stockarton Moor). Sulphur isotopes indicate that magmatic-

1242 hydrothermal fluid exsolved and mixed with hydrothermal fluids derived from contact  
1243 metamorphic dewatering of the country rocks.

1244 In soft collision zones magma is considered to have an important role in the transfer  
1245 of energy (as heat) and mass to shallow crustal levels beyond the range traditionally indicated  
1246 for orogenic gold deposits. Post-subduction soft continental collisional orogenic systems are  
1247 considered important targets for gold mineralisation. This case study demonstrates that soft  
1248 continental collision zones are likely to be inherently prospective for mineralisation for five  
1249 reasons:

1250 1) Delamination of the lithospheric mantle from the crust of the down-going plate is  
1251 considered likely to occur during soft continental collision due to the relatively slow rate of  
1252 advance of the down-going plate relative to the rate of descent due to the negative buoyancy  
1253 of the slab.

1254 2) The previous history of subduction of Iapetus metasomatically hydrated and  
1255 fertilised the lithospheric mantle. A fertile source region is a critical element of a mineralising  
1256 system and is clearly inherent in collisional orogenic belts.

1257 3) Transient geodynamics, for example the change in deformation regime from  
1258 orthogonal to strike-slip, are common during soft continental collision and are recognised as  
1259 a critical element of mineralising systems.

1260 4) The switch to transtension provides a favourable lithospheric architecture for the  
1261 effective and rapid flux of mass and energy from deep to shallow environments.

1262 5) Continental crust is unlikely to be subjected to significant tectonic thickening during  
1263 soft collision, resulting in low degrees of subsequent exhumation and an increased  
1264 preservation potential for high-level mineral deposits.

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1268     of the West of Scotland.  
1269  
1270

1271 Figure 1. Map of the Southern Uplands-Down-Longford Terrane showing features and  
 1272 locations referred to in the text: BS: Black Stockarton; CL: Clay Lake; CT: Clontibret; CW:  
 1273 Conlig-Whitespots; FB: Fore Burn; GD: Glendinning; GH: Glenhead; GI: Glenish; HH: Hare Hill;  
 1274 LW: Leadhills-Wanlockhead; MH: Moorbrock Hill; SG: Slieve Glah; TN: Talnotry. Permo-  
 1275 Carboniferous volcano-sedimentary basins: 1: Kinscourt; 2: Strangford; 3: North Channel; 4:  
 1276 Stranraer/Luce; 5: Thornhill; 6: Dumfries; 7: Langholm. Caledonian plutonic complexes: a:  
 1277 Crossdoney; b: Newry; c: Loch Doon; d: Fleet; e: Cairnsmore of Carsphairn; f: Criffel-Dalbeattie.  
 1278 Named structures: CF: Cloughy Fault; LF: Laurieston Fault; MSZ/SGSZ: Moniaive Shear Zone  
 1279 and Slieve Glah Shear Zone; OBF: Orlock Bridge Fault. Based upon data provided by the British  
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1281  
 1282 Figure 2. Schematic block diagram showing generalised structural and tectonostratigraphic  
 1283 relationships within the SUDLT based on the work of Anderson (2001) and Anderson and  
 1284 Cameron (1979).

1285  
 1286 Figure 3. Structural data. a) poles to veins at Hare Hill after Boast et al. (1990); b) strike  
 1287 orientations of dextral and sinistral D<sub>3</sub> strike-slip faults, Ards peninsula, Co. Down after  
 1288 Anderson et al. (1995); c) strike orientations of dextral and sinistral D<sub>3</sub> strike-slip faults, Rhinns  
 1289 of Galloway after Stone (1995); d) strike orientations of dextral and sinistral D<sub>3</sub> strike-slip  
 1290 faults, Wigtownshire after Barnes (2008); e) strike orientations of Pb-Zn veins at Leadhills-  
 1291 Wanlockhead after Temple (1957); f) strike orientations of fractures at Glenhead after Leake  
 1292 et al., 1981.

1293  
 1294 Figure 4. Structural control of geochemical anomalies. a) Au in bedrock at Hare Hill showing  
 1295 a dominant NE-SW Caledonoid pattern with weaker intersecting ~N-S anomalies (Boast et al.,  
 1296 1990). b) arsenic anomaly map of Glenhead Burn showing concordant NE-SW anomalies  
 1297 intersected by a ~N-S discordant trend (Leake et al., 1981).

1298

1299 Figure 5. Compositions of lamprophyric rocks of the SUDLT plotted on a hierarchical series  
1300 of geochemical tectonic discrimination diagrams for potassic igneous rocks following Muller  
1301 and Groves (2016). Geochemical data provided by BGS under licence IPR/191-244DX. Fields  
1302 for potassic igneous rocks: CAP: continental arc; IOP: initial oceanic arc; LOP: late oceanic  
1303 arc; PAP: post-collisional arc; WIP: within-plate. Based upon data provided by the British  
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1305

1306 Figure 6. Schematic representations of zoned alteration mineral assemblages at Glenhead,  
1307 Clontibret and Black Stockarton Moor based on descriptions in Leake et al. (1981), Brown et  
1308 al. (1979) and Morris (1984).

1309

1310 Figure 7. Schematic map showing inferred cross-cutting relationships between country rocks,  
1311 major and minor intrusions, alteration and veining at Glenhead Burn based on descriptions in  
1312 Leake et al. (1981).

1313

1314 Figure 8. Geochemical plots of rock samples from Clontibret: a) As ppm vs K<sub>2</sub>O/Na<sub>2</sub>O, b) Sb  
1315 ppm vs K<sub>2</sub>O/Na<sub>2</sub>O, c) K<sub>2</sub>O/Na<sub>2</sub>O 15m profile across the main lode zone, d) MgO/CaO 15m  
1316 profile across the main lode zone. After Morris et al. (1986).

1317

1318 Figure 9. Comparison of ranges of homogenisation temperatures and salinity for fluid  
1319 inclusions in early (Caledonian) auriferous quartz veins and late Pb-Zn sulphide veins in the  
1320 SUDLT. Early veins: 1: Glendinning (Duller et al., 1997); 2: Clontibret (Steed and Morris, 1986);  
1321 3: Conlig-Whitespots and Castleward (Baron and Parnell, 2000); 4: Black Stockarton Moor  
1322 (Lowry et al., 1997); 5: Leadhills-Wanlockhead (Samson and Banks, 1988); 6: Hare Hill (Samson  
1323 and Banks, 1988); Late Pb-Zn veins: 7: Conlig-Whitespots and Castleward (Baron and Parnell,  
1324 2000); 8: Black Stockarton Moor (Lowry et al., 1997); 9: Southern Uplands (Leadhills-  
1325 Wanlockhead, Woodhead, Hare Hill, Coldstream Burn, Blackcraig, Pibble and Enrick; Samson  
1326 and Banks, 1988).

1327

1328 Figure 10. Histograms showing homogenisation temperature data for fluid inclusions from  
 1329 'early' (Caledonian) quartz veins and later Pb-Zn carbonate veins at Leadhills-Wanlockhead  
 1330 after Samson and Banks (1988).

1331

1332 Figure 11. Comparison of  $\delta^{34}\text{S}$  for sulphides for Caledonian gold-bearing mineralised localities  
 1333 in the SUDLT. Also shown are Leadhills Pb-Zn veins together with diagenetic pyrite from shale  
 1334 (Moffat Shale; MFS) at Leadhills and Clontibret. Data from Lowry (1992), Steed and Morris  
 1335 (1997), Samson and Banks (1988), Duller et al. (1997), Anderson et al. (1989).

1336

1337 Figure 12. Oxygen-hydrogen isotope relationships for mineralising fluids for Clontibret lode  
 1338 gold (after Steed and Morris, 1997), Talnotry and Cairngarroch Bay (after Lowry et al., 1997)  
 1339 and Pb-Zn carbonate veins at Leadhills (after Samson and Banks, 1988).  $\delta\text{D}$  values for Leadhills  
 1340 from fluid inclusions.  $\delta^{18}\text{O}$  values calculated from mineral values using fractionation factors  
 1341 and temperatures of 80-120°C. Magmatic and meteoric water compositions from Taylor  
 1342 (1979). Meteoric water line from Craig (1961).

1343

1344 Figure 13. a) Contoured map of metamorphic grade in the central and western Southern  
 1345 Uplands, south-west Scotland after Merriman and Roberts (2000). b) map of arsenic  
 1346 abundances in stream sediments (BGS G-base geochemical survey data ©NERC)  
 1347 interpolated using Kriging. SUF: Southern Uplands Fault. OBF: Orlock Bridge Fault. Basins  
 1348 and plutons shown as for (a).

1349

1350

1351 Figure 14. Preferred model for the geodynamic setting and regional scale controls of  
 1352 Caledonian gold mineralisation in the SUDLT (after Miles et al., 2016). 'SCLM: sub-  
 1353 continental lithospheric mantle.

1354

1355 Table 1: Paragenetic sequence at Clontibret (after Morris, 1984).

1356

1357 Table 2: Summary of gold occurrences in the SUDLT.

1358

1359 Table 3: Summary of fluid inclusion data.

1360

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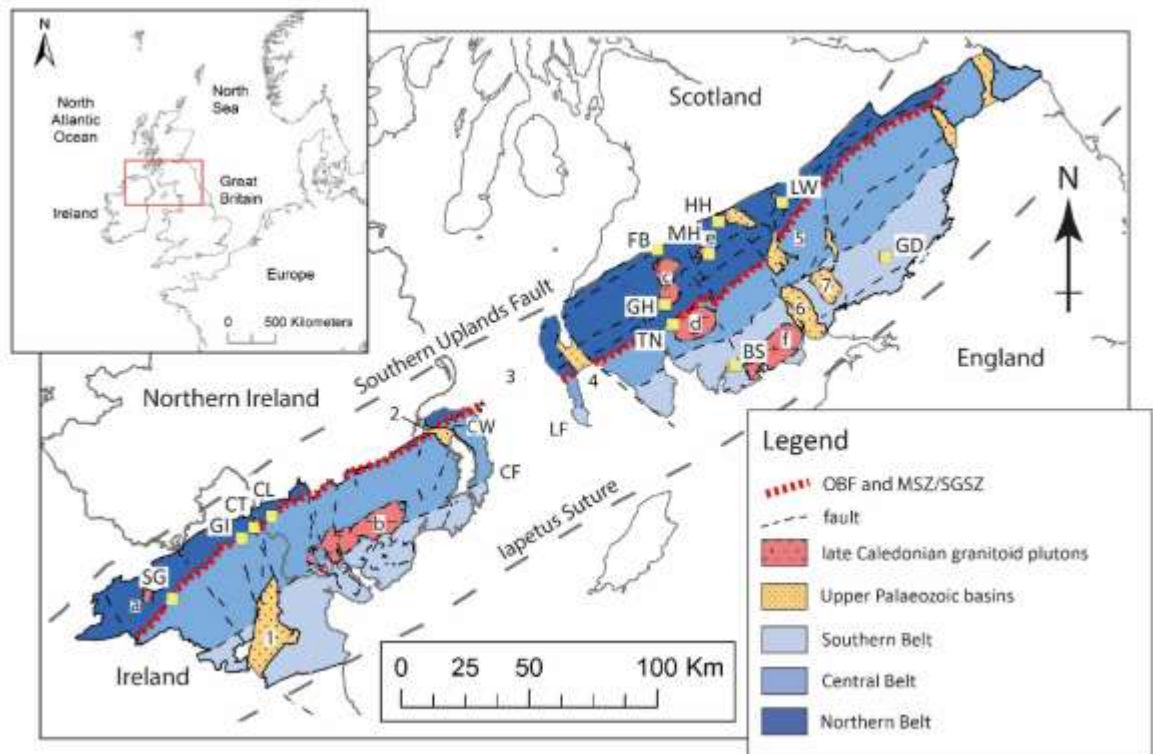
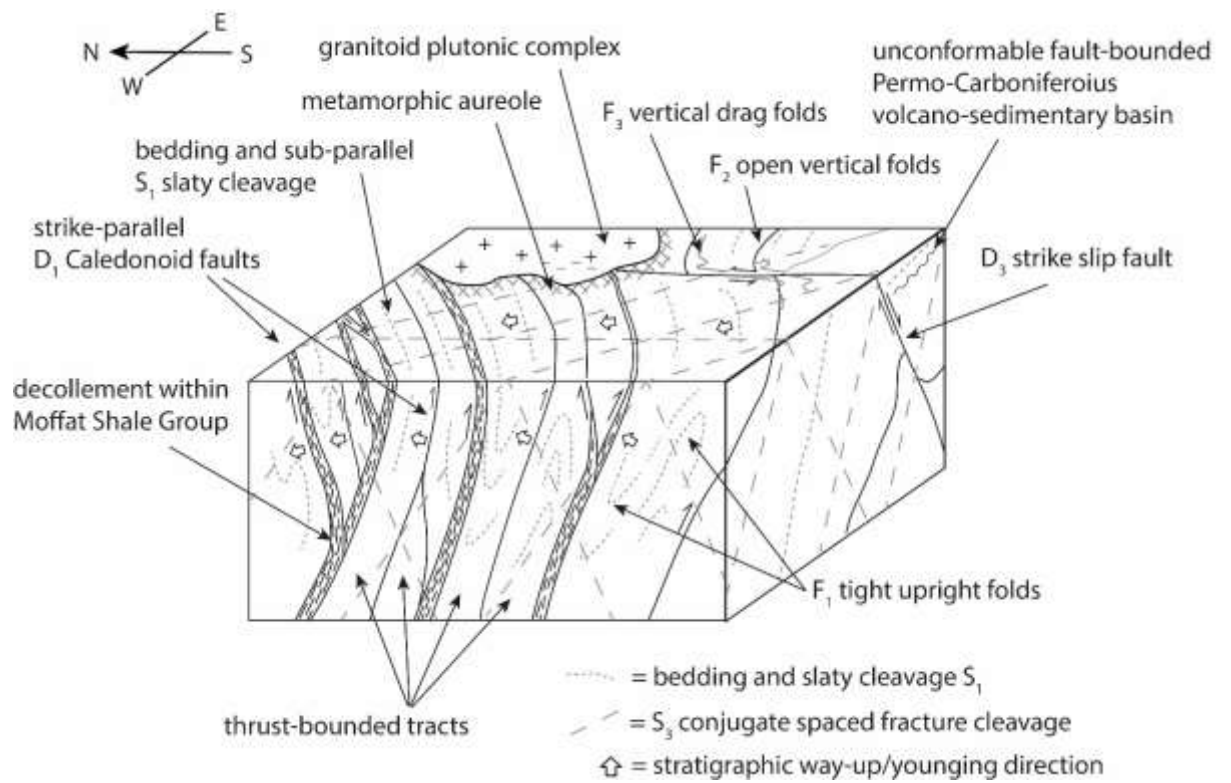


Figure 1. Map of the Southern Uplands-Down-Longford Terrane showing features and locations referred to in the text: BS: Black Stockarton; CL: Clay Lake; CT: Clontibret; CW: Conlig-Whitespots; FB: Fore Burn; GD: Glendinning; GH: Glenhead; GI: Glenish; HH: Hare Hill; LW: Leadhills-Wanlockhead; MH: Moorbrock Hill; SG: Slieve Glah; TN: Talnotry. Permo-Carboniferous volcano-sedimentary basins: 1: Kinscourt; 2: Strangford; 3: North Channel; 4: Stranraer/Luce; 5: Thornhill; 6: Dumfries; 7: Langholm. Caledonian plutonic complexes: a: Crossdoney; b: Newry; c: Loch Doon; d: Fleet; e: Cairnsmore of Carsphairn; f: Criffel-Dalbeattie. Named structures: CF: Cloughy Fault; LF: Laurieston Fault; MSZ/SGSZ: Moniaive Shear Zone and Slieve Glah Shear Zone; OBF: Orlock Bridge Fault. Based upon data provided by the British Geological Survey - Licence No. 2015/162 ED © NERC. All rights reserved.



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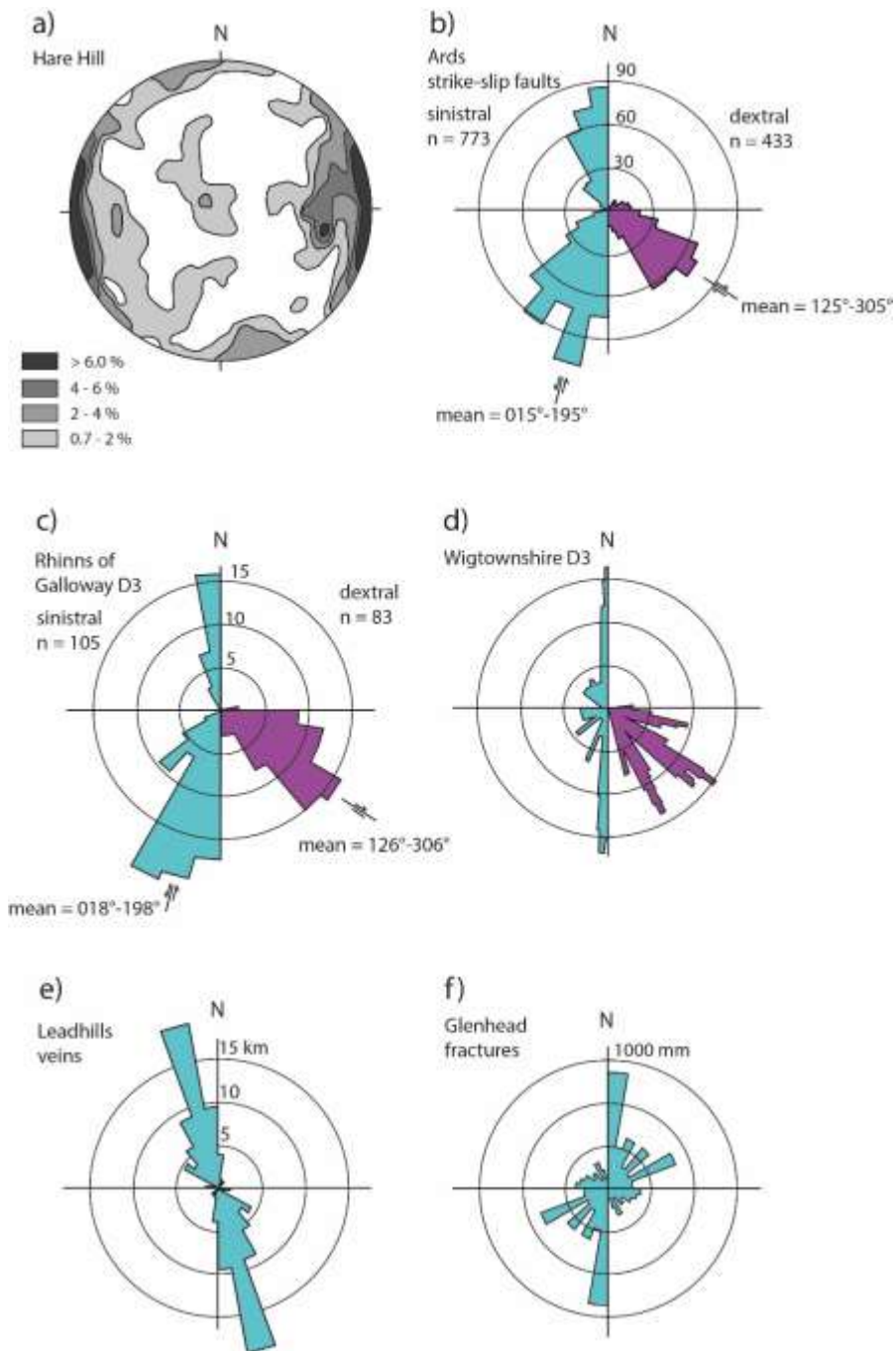


15

16 Figure 2. Schematic block diagram showing generalised structural and tectonostratigraphic  
 17 relationships within the SUDLT based on the work of Anderson (2001) and Anderson and  
 18 Cameron (1979).

19





21

22 Figure 3. Structural data. a) poles to veins at Hare Hill after Boast et al. (1990); b) strike  
 23 orientations of dextral and sinistral D<sub>3</sub> strike-slip faults, Ards peninsula, Co. Down after  
 24 Anderson et al. (1995); c) strike orientations of dextral and sinistral D<sub>3</sub> strike-slip faults, Rhinns  
 25 of Galloway after Stone (1995); d) strike orientations of dextral and sinistral D<sub>3</sub> strike-slip  
 26 faults, Wigtownshire after Barnes (2008); e) strike orientations of Pb-Zn veins at Leadhills-  
 27 Wanlockhead after Temple (1957); f) strike orientations of fractures at Glenhead after Leake  
 28 et al., 1981.



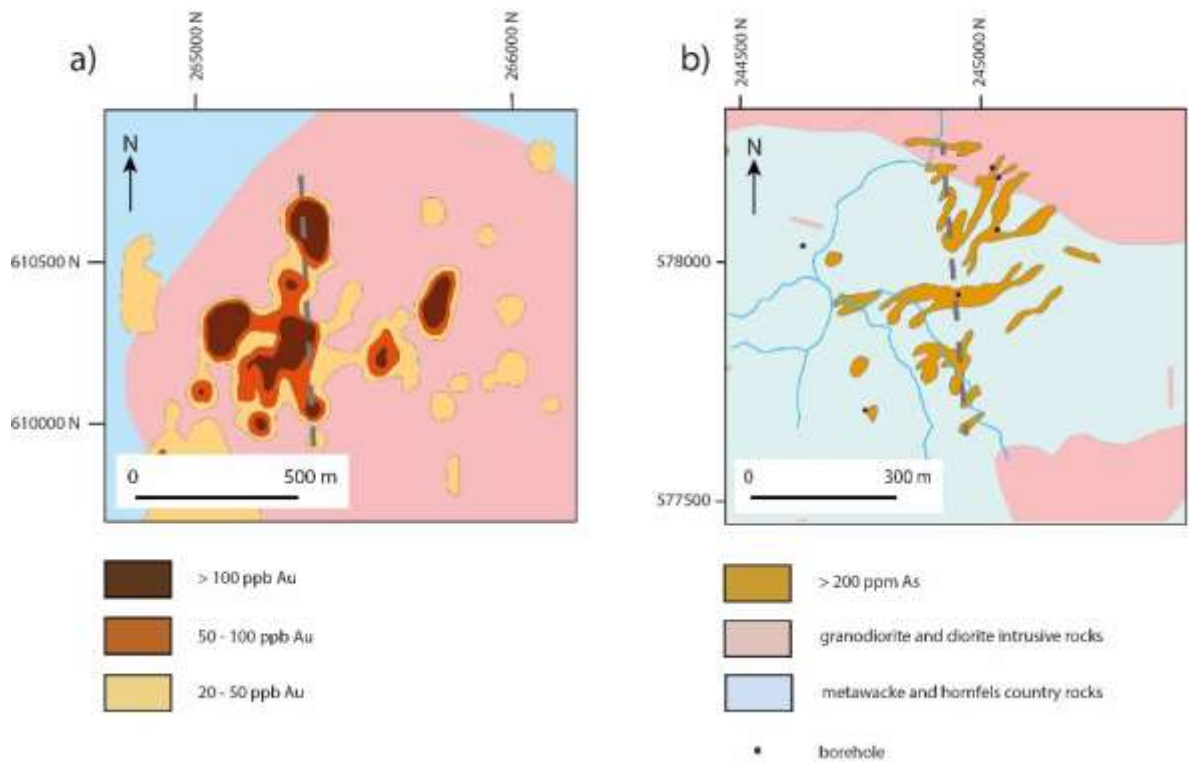
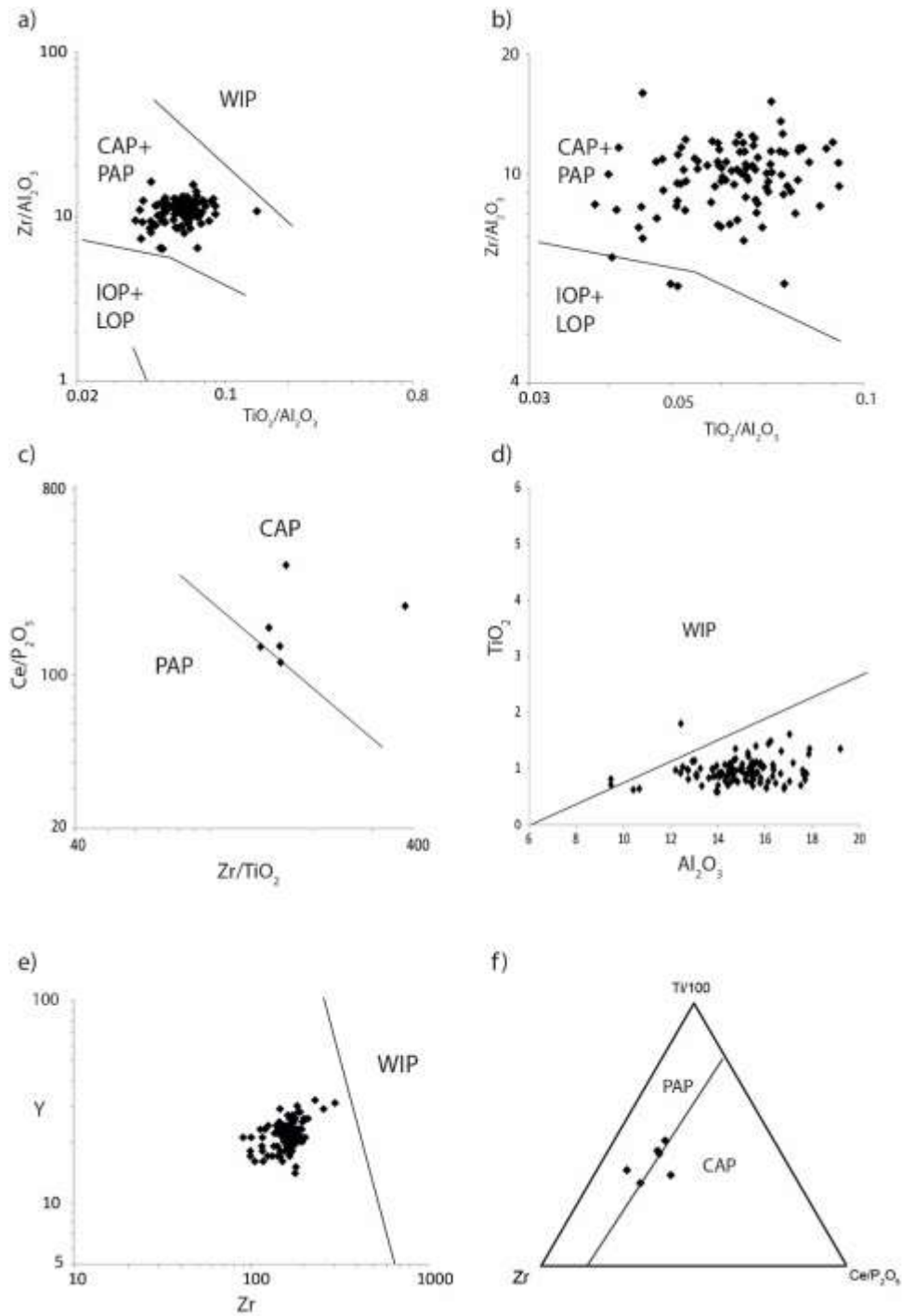


Figure 4. Structural control of geochemical anomalies. a) Au in bedrock at Hare Hill showing a dominant NE-SW Caledonoid pattern with weaker intersecting ~N-S anomalies (Boast et al., 1990). b) arsenic anomaly map of Glenhead Burn showing concordant NE-SW anomalies intersected by a ~N-S discordant trend (Leake et al., 1981).







37

38 Figure 5. Compositions of lamprophyric rocks of the SUDLT plotted on a hierarchical series  
 39 of geochemical tectonic discrimination diagrams for potassic igneous rocks following Muller  
 40 and Groves (2016). Geochemical data provided by BGS under licence IPR/191-244DX. Fields  
 41 for potassic igneous rocks: CAP: continental arc; IOP: initial oceanic arc; LOP: late oceanic  
 42 arc; PAP: post-collisional arc; WIP: within-plate. Based upon data provided by the British  
 43 Geological Survey © NERC. All rights reserved.



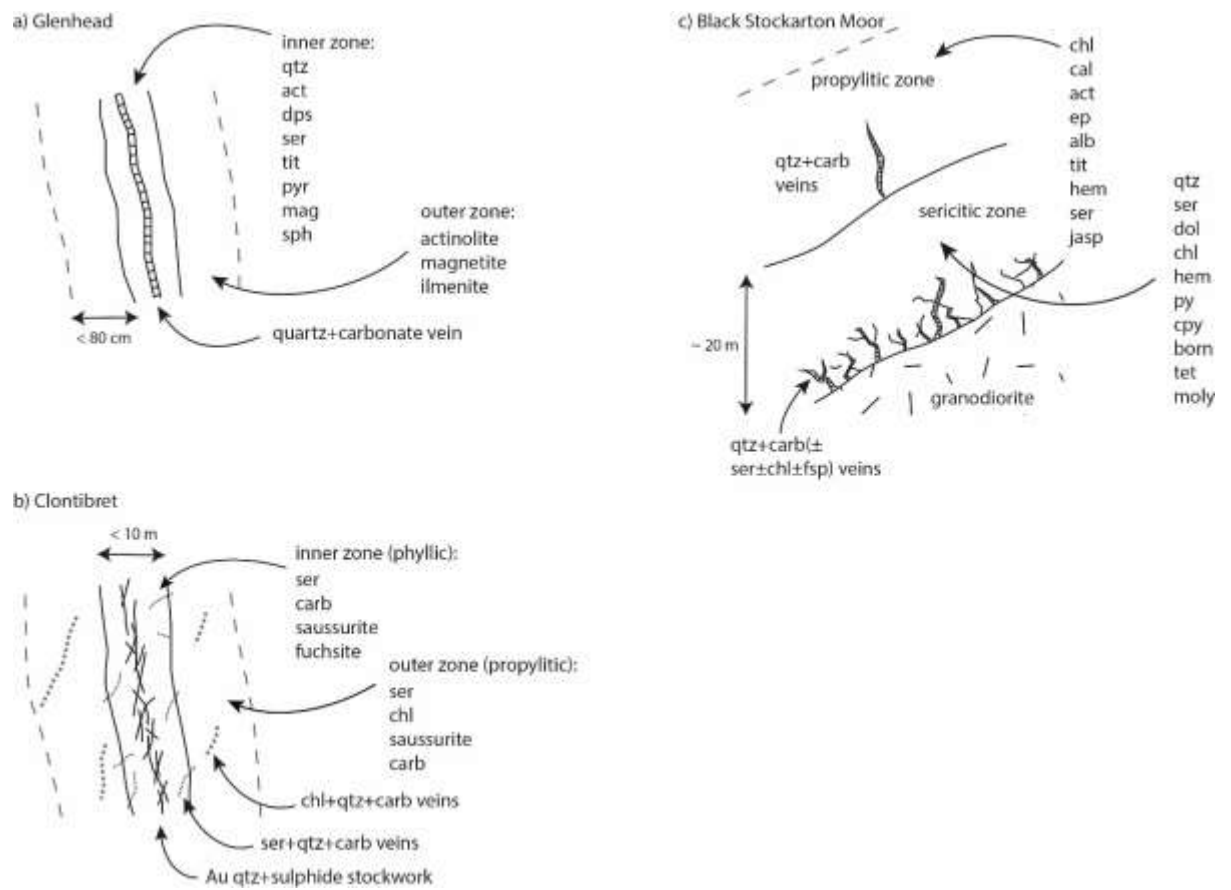
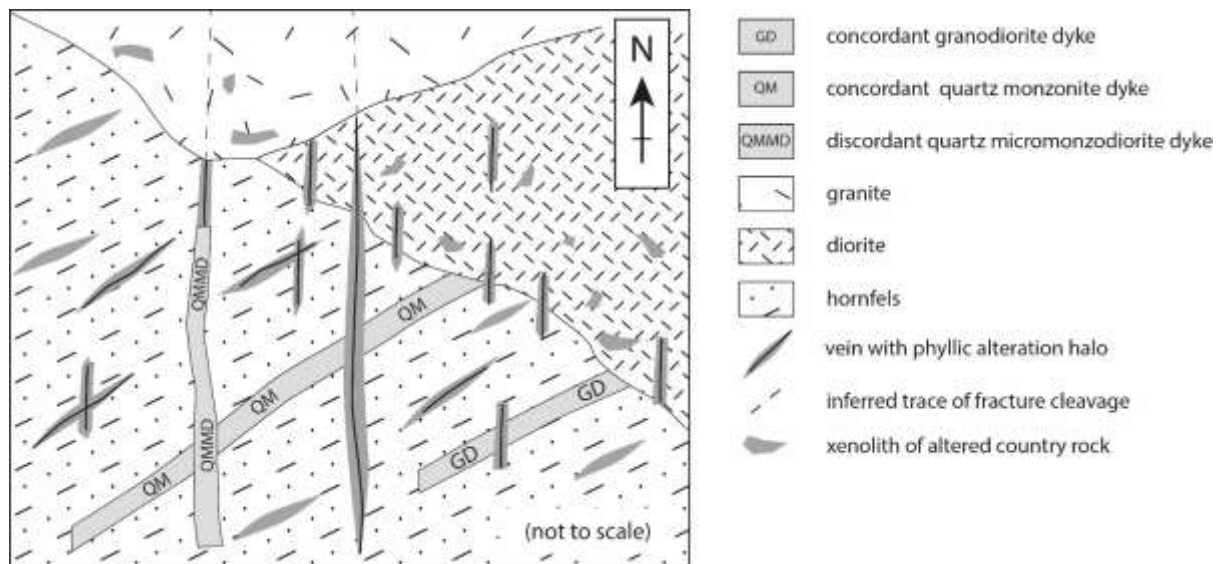


Figure 6. Schematic representations of zoned alteration mineral assemblages at Glenhead, Clontibret and Black Stockarton Moor based on descriptions in Leake et al. (1981), Brown et al. (1979) and Morris (1984).





51  
 52 Figure 7. Schematic map showing inferred cross-cutting relationships between country rocks,  
 53 major and minor intrusions, alteration and veining at Glenhead Burn based on descriptions in  
 54 Leake et al. (1981).



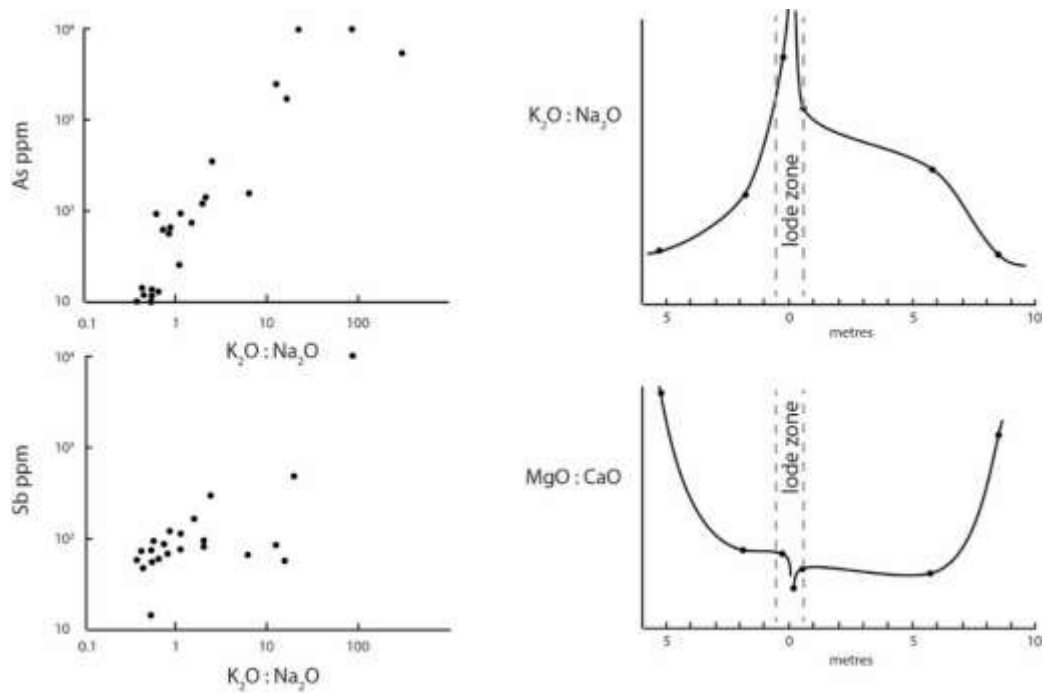


Figure 8. Geochemical plots of rock samples from Clontibret: a) As ppm vs  $K_2O/Na_2O$ , b) Sb ppm vs  $K_2O/Na_2O$ , c)  $K_2O/Na_2O$  15m profile across the main lode zone, d)  $MgO/CaO$  15m profile across the main lode zone. After Morris et al. (1986).





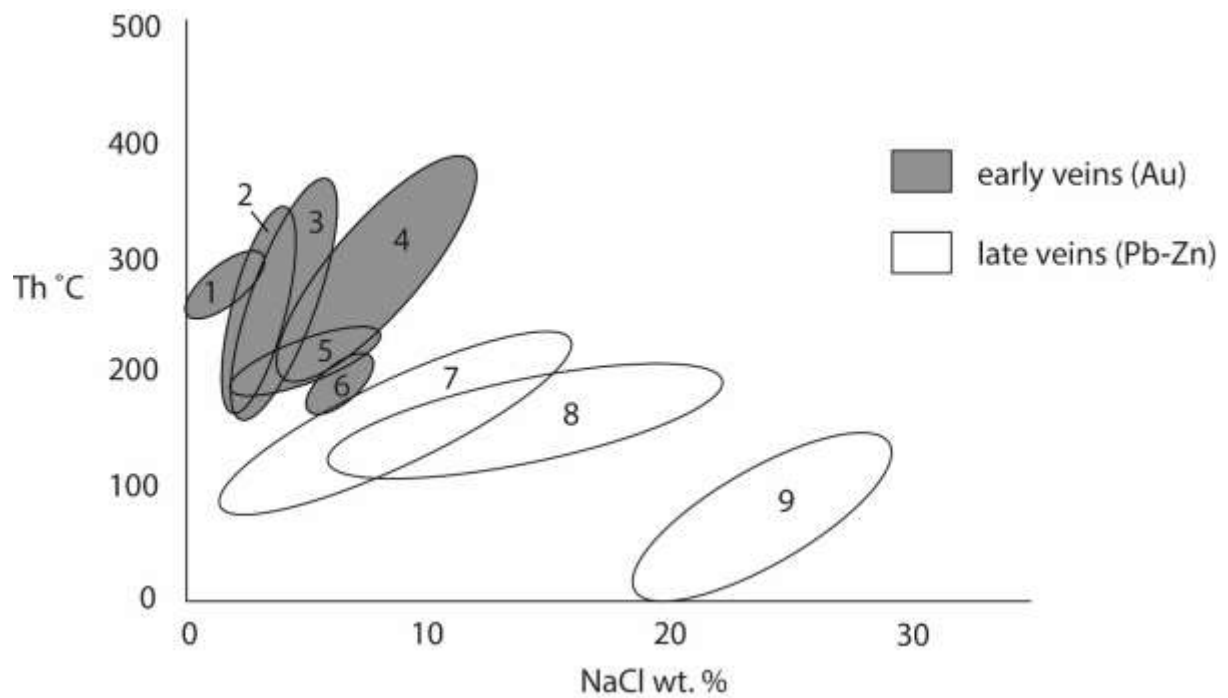


Figure 9. Comparison of ranges of homogenisation temperatures and salinity for fluid inclusions in early (Caledonian) auriferous quartz veins and late Pb-Zn sulphide veins in the SUDLT. Early veins: 1: Glendinning (Duller et al., 1997); 2: Clontibret (Steed and Morris, 1986); 3: Conlig-Whitespots and Castleward (Baron and Parnell, 2000); 4: Black Stockarton Moor (Lowry et al., 1997); 5: Leadhills-Wanlockhead (Samson and Banks, 1988); 6: Hare Hill (Samson and Banks, 1988); Late Pb-Zn veins: 7: Conlig-Whitespots and Castleward (Baron and Parnell, 2000); 8: Black Stockarton Moor (Lowry et al., 1997); 9: Southern Uplands (Leadhills-Wanlockhead, Woodhead, Hare Hill, Coldstream Burn, Blackcraig, Pibble and Enrick; Samson and Banks, 1988).



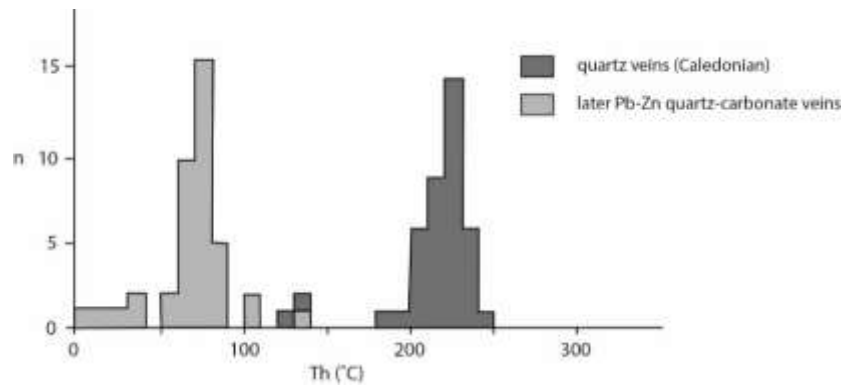


Figure 10. Histograms showing homogenisation temperature data for fluid inclusions from 'early' (Caledonian) quartz veins and later Pb-Zn carbonate veins at Leadhills-Wanlockhead after Samson and Banks (1988).



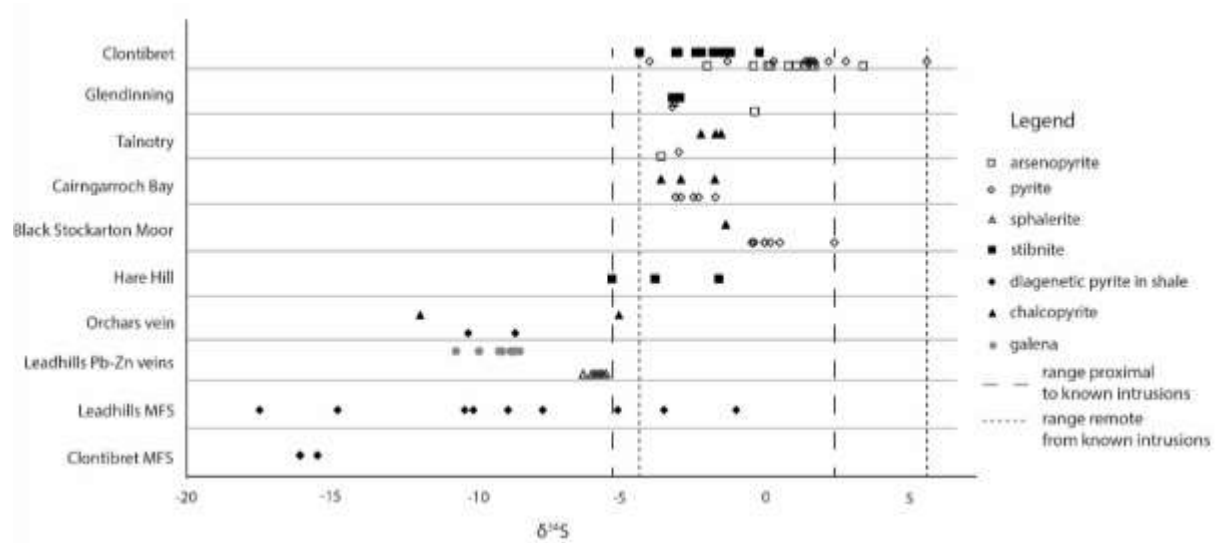


Figure 11. Comparison of  $\delta^{34}\text{S}$  for sulphides for Caledonian gold-bearing mineralised localities in the SUDLT. Also shown are Leadhills Pb-Zn veins together with diagenetic pyrite from shale (Moffat Shale; MFS) at Leadhills and Clontibret. Data from Lowry (1992), Steed and Morris (1997), Samson and Banks (1988), Duller et al. (1997), Anderson et al. (1989).



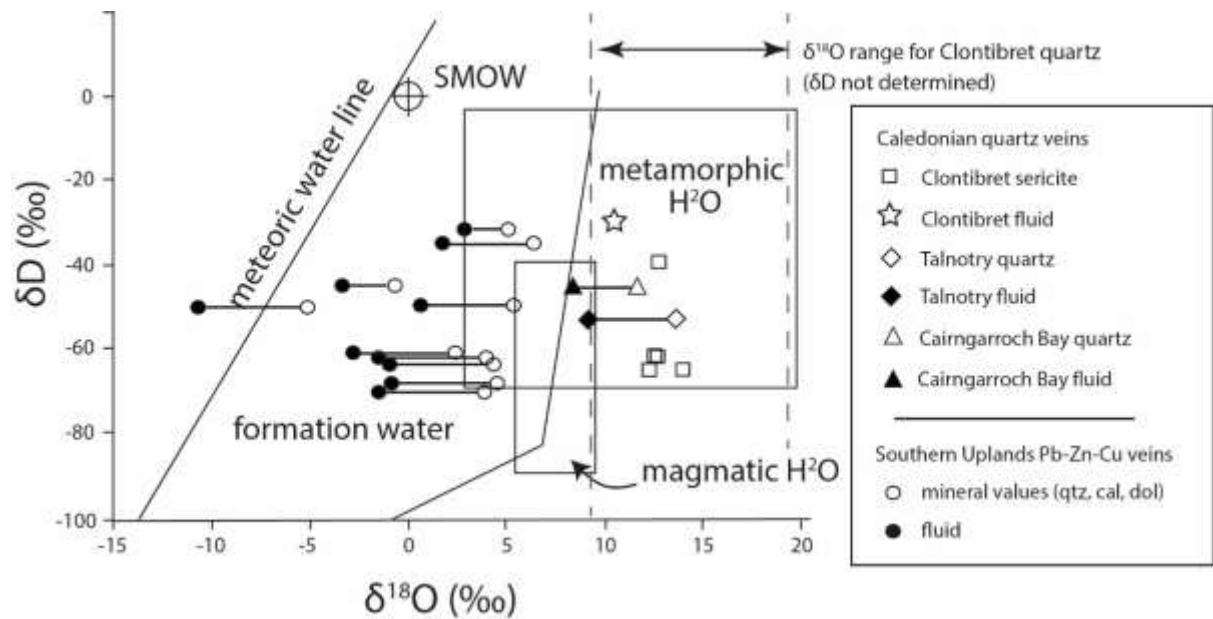
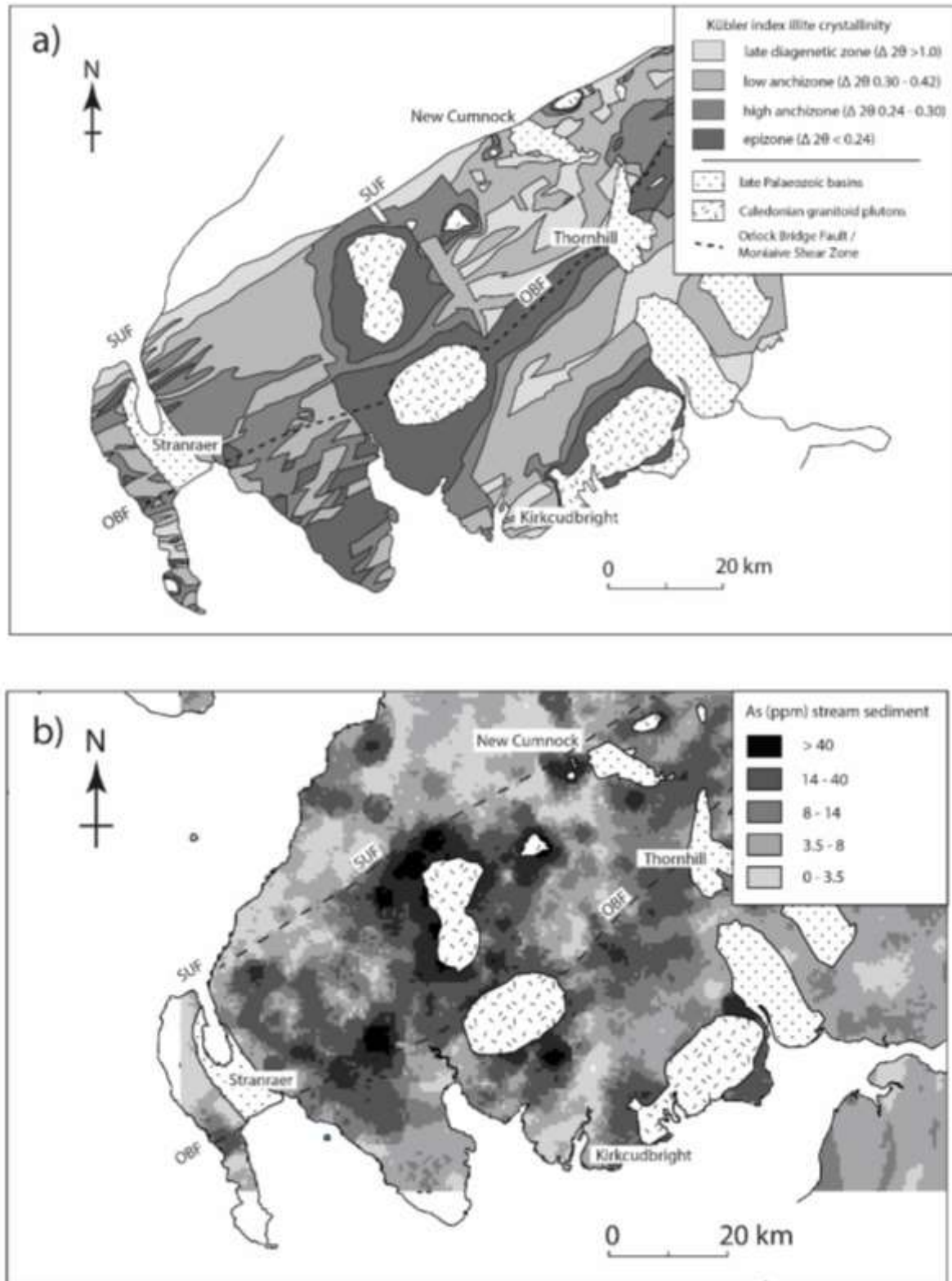


Figure 12. Oxygen-hydrogen isotope relationships for mineralising fluids for Clontibret lode gold (after Steed and Morris, 1997), Talnotry and Cairngarroch Bay (after Lowry et al., 1997) and Pb-Zn carbonate veins at Leadhills (after Samson and Banks, 1988).  $\delta D$  values for Leadhills from fluid inclusions.  $\delta^{18}O$  values calculated from mineral values using fractionation factors and temperatures of 80-120°C. Magmatic and meteoric water compositions from Taylor (1979). Meteoric water line from Craig (1961).







98

99 Figure 13. a) Contoured map of metamorphic grade in the central and western Southern

100 Uplands, south-west Scotland after Merriman and Roberts (2000). b) map of arsenic

101 abundances in stream sediments (BGS G-base geochemical survey data ©NERC)

102 interpolated using Kriging. SUF: Southern Uplands Fault. OBF: Orlock Bridge Fault. Basins  
103 and plutons shown as for (a).  
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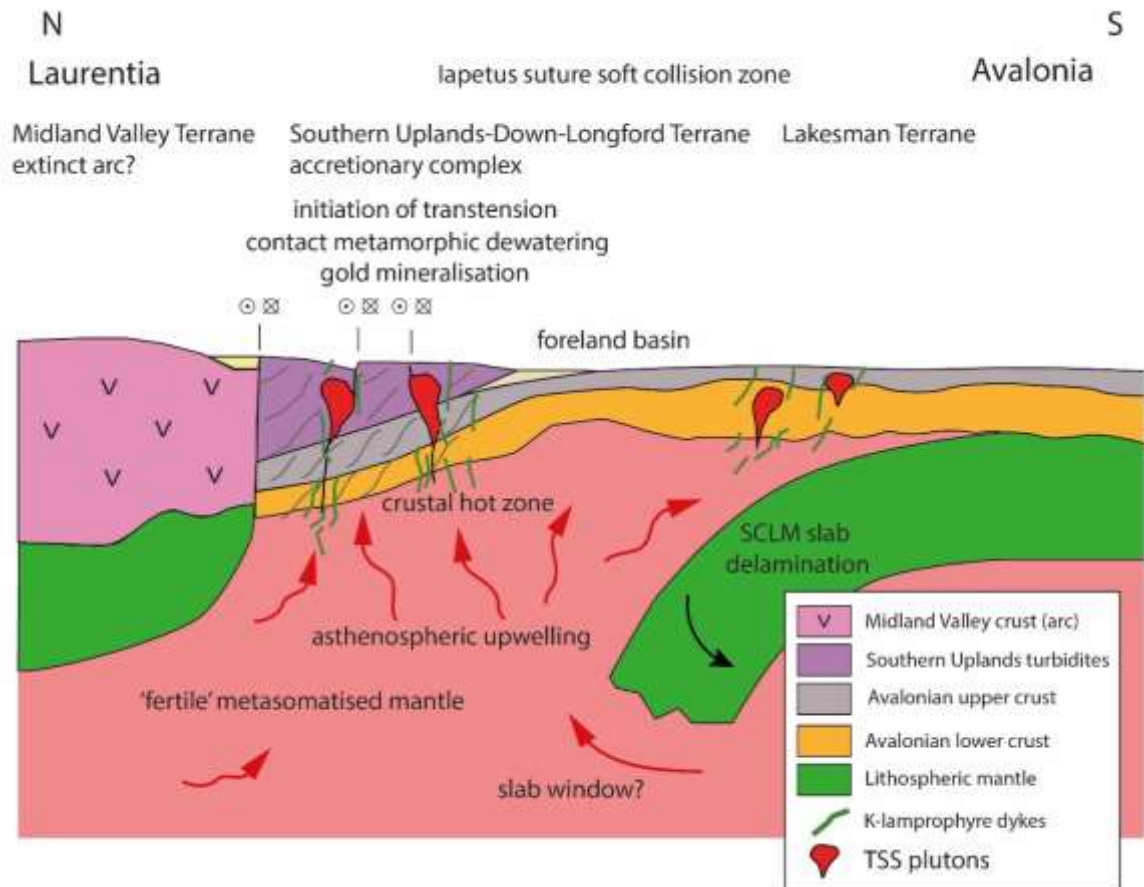


Figure 14. Preferred model for the geodynamic setting and regional scale controls of Caledonian gold mineralisation in the SUDLT (after Miles et al., 2016). 'SCLM: sub-continental lithospheric mantle.



Table 1: Paragenetic sequence at Clontibret (after Morris, 1984).

[illegible]



Table 2: Summary of gold occurrences in the SUDLT.

deposit name	Au max (ppm)	lithology	stratigraphy	lode control	major structure
Slieve Glah	1.7	Black Shale	(equivalent to Moffat shale)	unknown	Orlock Bridge Fault
Glenish	9.4	Turbidites	Shinnel Formation	unknown	Orlock Bridge Fault
Clontibret	2.5m @ 25	Turbidites	equivalent to Gala Group	N-S	Orlock Bridge Fault
Clay Lake	5m @ 3.02	Black Shale	(equivalent to Moffat shale)	unknown	Orlock Bridge Fault
Fore Burn	0.25m @ 52	granodiorite	n/a	NW-SE	Southern Uplands Fault
Moorbrock Hill	10m @ 4.85	diorite and black shale	Moffat shale	N-S, NE-SW	Leadhills Fault
Glenhead burn	1m @ 8.8	diorite and turbidites	Glenwhargen Formation	N-S, NE-SW	Fardingmullach Fault
Black Stockarton Moor	0.06	turbidites	Hawick Group	unknown	none
Leadhills	0.4	turbidites	Portpatrick Formation	unknown	Leadhills Fault
Glendinning	0.84	turbidites	Hawick Group	N-S	Lauriestoun Fault
Duns	5	turbidites	Gala	unknown	Leadhills Fault
Hare Hill		granodiorite	n/a	N-S, NE-SW	none





Table 3: Summary of fluid inclusion data.

early veins	NaCl min	NaCl max	t min	t max	source
Clontibret	2	4	170	340	Steed and Morris, 1986
Glendinning	0	3	250	300	Duller et al., 1997
Leadhills	2	8	187	236	Samson and Banks, 1988
Black Stockarton	4.4	11.7	197	386	Lowry et al., 1997
Hare Hill	5.2	7.6	168	213	Samson and Banks, 1988
Castleward and Conlig	2.41	5.86	158	367	Baron and Parnell, 2000
late veins					
Southern Uplands	19	29	5	134	Samson and Banks, 1988
Black Stockarton	6	22	123	188	Lowry et al., 1997
Castleward and Conlig	1.4	15.86	83	228	Baron and Parnell, 2000





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Clay Lake	5m @ 3.02	Black Shale	(equivalent to Moffat shale)	unknown	Southern Uplands Fault
Fore Burn	0.25m @ 52	granodiorite diorite and	n/a	NW-SE	Fault
Moorbrock Hill	10m @ 4.85	black shale diorite and	Moffat shale Glenwhargen	N-S, NE-SW	Leadhills Fault Fardingmullach
Glenhead burn	1m @ 8.8	turbidites	Formation	N-S, NE-SW	Fault
Black Stockarton Moor	0.06	turbidites	Hawick Group Portpatrick	unknown	none
Leadhills	0.4	turbidites	Formation	unknown	Leadhills Fault
Glendinning	0.84	turbidites	Hawick Group	N-S	Lauriestoun Fault
Duns	5	turbidites	Gala	unknown	Leadhills Fault
Hare Hill		granodiorite	n/a	N-S, NE-SW	none



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